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TEMPERATURE OSCILLATIONS INVOLVING
THE UNSATURATED HELIUM FILM

A THESIS

SUBMITTED TO THE FACULTY OF GRADUATE STUDIES
IN PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE
OF MASTER OF SCIENCE .

DEPARTMENT OF PHYSICS

by

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The undersigned certify that they have read,
and recommend to the Faculty of Graduate Studies for
acceptance, a thesis entitled Temperature Oscillations
Involving the Unsaturated Helium Film, submitted by
Janette Adele Buckley in partial fulfilment of the
requirements for the degree of Master of Science.

TABLE OF CONTENTS

	<u>Page</u>
ACKNOWLEDGEMENTS	
ABSTRACT	
INTRODUCTION	1
I. HELIUM FILMS - SATURATED AND UNSATURATED	9
The Saturated Film	11
The Unsaturated Film and Helium Adsorption	14
Superfluidity in the Unsaturated Film . .	20
II. EXPERIMENTAL APPROACH AND THE APPARATUS	28
The Experimental Chamber	28
Modifications of the Experimental Capsules	35
The Helium Gas Filling System	36
General Features of the Demagnetization	
Cryostat	38
Pressure Measurements	43
Electrical Measurements	45
III. EXPERIMENTAL PROCEDURE	48
Preparation for an Experiment	48
Cooling by Adiabatic Demagnetization . . .	49
The Observation of Temperature Oscillations	
due to Unsaturated Film Flow	53
IV. EXPERIMENTAL RESULTS	56
V. CONCLUSIONS	85
APPENDIX I	
The Carbon Resistance Thermometers	92
APPENDIX II	
The Oil McLeod Pressure Gauge	97
REFERENCES	100

LIST OF ILLUSTRATIONS

	<u>Page</u>
Fig. 1 Schematic diagram of experimental chamber and temperature oscillations	6
Fig. 2 Onset of superfluidity as a function of relative saturation pressure and temperature	23
Fig. 3 Heat transport apparatus used by Long and Meyer	23
Fig. 4 Photograph of experimental chamber	30
Fig. 5 The helium gas filling system	37
Fig. 6 Low temperature section of the apparatus	40
Fig. 7 Reproduction of the direct observation of temperature oscillations	60
Fig. 8 The effect of several types of thermal contact on can temperature	61
Fig. 9 Onset of superfluidity as a function of helium input	68
Fig.10 The onset of superflow as indicated by a change of warming rate of the can	70
Fig.11 Dependence of the base line temperature of the oscillations on film distribution	72
Fig.12 Dependence of average oscillation amplitude on the unsaturated film thickness	74
Fig.13 Time occupied by a set of oscillations vs. helium input	76
Fig.14 Filling system pressure vs. time	82
Fig.15 An illustration of the sharpness of the film transition	91
Fig.16 The oil McLeod pressure gauge	98

LIST OF TABLES

		Page
Table I	The effect on oscillation amplitude of changing the initial magnetic field . .	77
Table II	A summary of amplitude changes caused by thermal switching	80
Table III	Coefficients calculated for thermometer calibration	94
Table IV	Oil McLeod calibration	99

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ABSTRACT

This thesis is the description of an experiment in which temperature oscillations of an experimental sample occurred as a result of heat transported by the adsorption and desorption of an unsaturated helium film. Oscillations of a similar nature were observed in earlier work by other investigators (Mendoza, 1948; Dugdale and MacDonald, 1957), and reported as "anomalous" thermal behavior. The present experiment has been directed toward explaining this behavior and producing the temperature oscillations under known, controllable conditions.

The unsaturated film was formed in a closed system, at a temperature below 1°K , consisting of a copper canister of small heat capacity joined by a metallic tube of low thermal conductivity to a paramagnetic salt capsule, which served as a heat sink after being adiabatically demagnetized. When oscillations take place, the unsaturated film is present on what may be viewed as two connected surfaces between which a temperature gradient exists. Superfluid film flows toward the warmer region, releases its heat of adsorption there, thus raising the temperature of the warm surface above that corresponding

to the "onset" of superfluidity. Then the recently deposited film is desorbed from the warmer surface, thus cooling it, "pumped" as gas to condense on the cooler surface, and the cycle repeats.

In addition to providing a satisfactory explanation of the poorly understood thermal behavior observed earlier, the present work has introduced some new aspects of the properties of the unsaturated helium film in that the "onset" of superfluidity was observed at temperatures as low as 0.6°K , where the transition was sharp to within 0.0002°K .

INTRODUCTION

This thesis describes experiments undertaken with the aim of understanding some unusual thermal behavior which has been observed from time to time during adiabatic demagnetization experiments in which a paramagnetic salt has been joined to an experimental sample by a metallic link. An explanation has been proposed and an experiment designed with the hope of producing similar thermal behavior under controlled conditions. The present work has been directed toward testing the possibility of temperature oscillations of a metallic sample occurring due to flow of an unsaturated helium film. Such oscillations have been produced in this laboratory at temperatures less than 1°K , with an apparatus that may be basically outlined as a warm surface connected by a poorly conducting metallic link to a cold surface. If a helium film is produced on these surfaces, it has been found that temperature oscillations of the warm body occur spontaneously under correct conditions of pressure, temperature gradient, and film thickness.

To introduce this work, a summary will be given of the several instances of unusual thermal behavior referred to above, always observed more or less

accidentally during the course of experiments involving adiabatic demagnetization, using helium gas for the purpose of heat exchange.

While investigating heat transport below 1°K , using two paramagnetic salt capsules, one "warm" and the other "cold", connected by a metal rod, Mendoza (1948) observed a temperature oscillation of the "warm" salt. Only about three complete cycles could be observed because of the excessive heat leak associated with this phenomenon. Since thorough pumping of the chamber surrounding the capsules caused a disappearance of the oscillations and a return to normal warming conditions after a demagnetization, Mendoza suggested the possibility that the oscillations were due, at least partially, to the presence of condensed helium on the capsules.

Dugdale and MacDonald (1957) observed oscillations in temperature of a sodium sample connected to a salt pill, while carrying out experiments on electrical resistance below 1°K . They also observed another unusual phenomenon which occurred in an experimental arrangement consisting of two salt capsules joined by a copper wire. They found that immediately heat was supplied to the warmer capsule, the temperature of the

cooler capsule decreased. In later experiments (Dugdale MacDonald and Croxon, 1957), they found these effects much less pronounced, and it was suggested that the possibility of the presence of an adsorbed helium film would be reduced by achieving a better insulating vacuum than in the previous work.

A similar effect to that just outlined has also been observed by Meyer (1959). His experimental arrangement consisted of several pieces of paramagnetic salt crystal, joined to a silver ribbon which had a heater at one end of it. When the heater was turned on, the salt crystals cooled at first, then warmed. In this experiment, it is of importance to note that Meyer relied on a procedure known as "baking-out" to produce satisfactory thermal insulation. "Baking-out" is described by Hull (1947) as follows. Helium condensed on the surface of a salt capsule will desorb as the capsule warms after demagnetization, thus causing deterioration of the high vacuum which is the means of thermal insulation of the capsule. To reduce this amount of condensed helium due to residual exchange gas, magnetization is carried out at, say, 1.5°K ; the exchange gas is pumped for a considerable length of time, and then the bath temperature is lowered to 1°K . In this

way, most of the remaining helium gas condenses on surfaces at 1°K , leaving the salt capsule relatively free of a helium film. Ideally, the effect of the "baking-out" process may be as described above, but since the lowering of the bath temperature is imperfectly adiabatic, a certain amount of thermal exchange is probably effected between salt and bath, and thus a considerable proportion of any residual helium gas may condense on the salt capsule in spite of these precautions. More helium gas, if present, may be expected to condense on the salt after demagnetization.

In each of the experiments just reviewed, it seemed reasonable to believe that a thin helium film may have been adsorbed on the surfaces of the bodies showing unusual thermal behavior. In addition, the observation of these phenomena seemed in some way associated with a temperature difference existing between parts of the experimental chamber joined by a metallic link. Further study was needed to discover if, and in what way, an adsorbed helium film was responsible for these effects. It was known that heat transport due to unsaturated film flow could occur, and that helium films existing at pressures less than the saturated vapor pressure for a given temperature had

an "onset" temperature for superfluid flow which continued to decrease below the bulk liquid lambda temperature as the relative saturation decreased. For details on these and other liquid helium properties, see Chapter I. On the basis of these properties together with the observed thermal effects, a mechanism for the temperature oscillations was proposed by Manchester, (Manchester and Buckley, 1960).

In a typical experimental arrangement, shown in the inset of Fig. 1, the system consists of a warmer body, B, joined by a metallic link, C, to a cooler body, A. (In the particular diagram shown, i.e. that used in this experiment, B is provided with a heater h. Both A and B have thermometers, t_1 and t_2 . A lead link between A and B supplies variable thermal contact, since it may be magnetically operated as a superconducting thermal switch. Helium gas is admitted through a capillary tube positioned above B.) A thin helium film is present on the surfaces of A, B, and C. When the salt capsule surrounding A is cooled by adiabatic demagnetization, the warmer body, B, will be cooled by conduction along C, until at some point on C, or B, the onset temperature for superfluid flow is passed. When this occurs, film flows toward B, i.e. up the temperature gradient. Consequently, the temperature

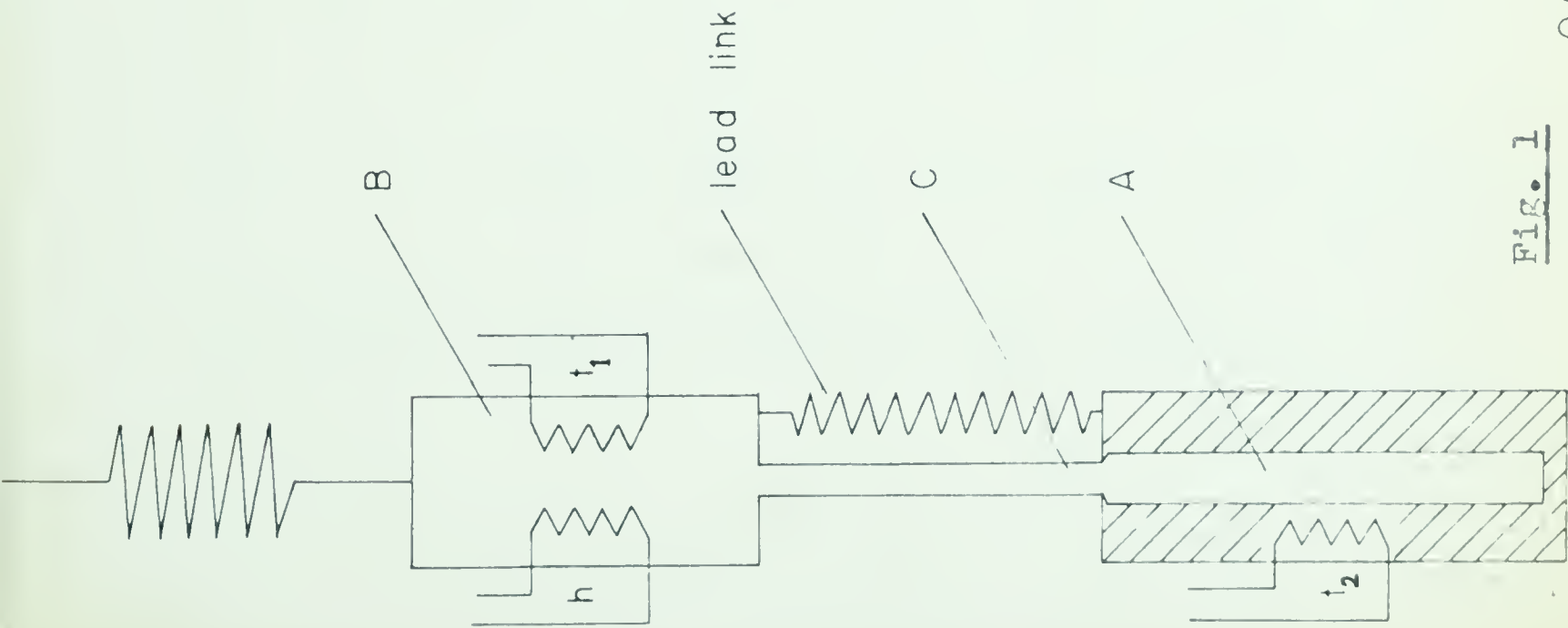
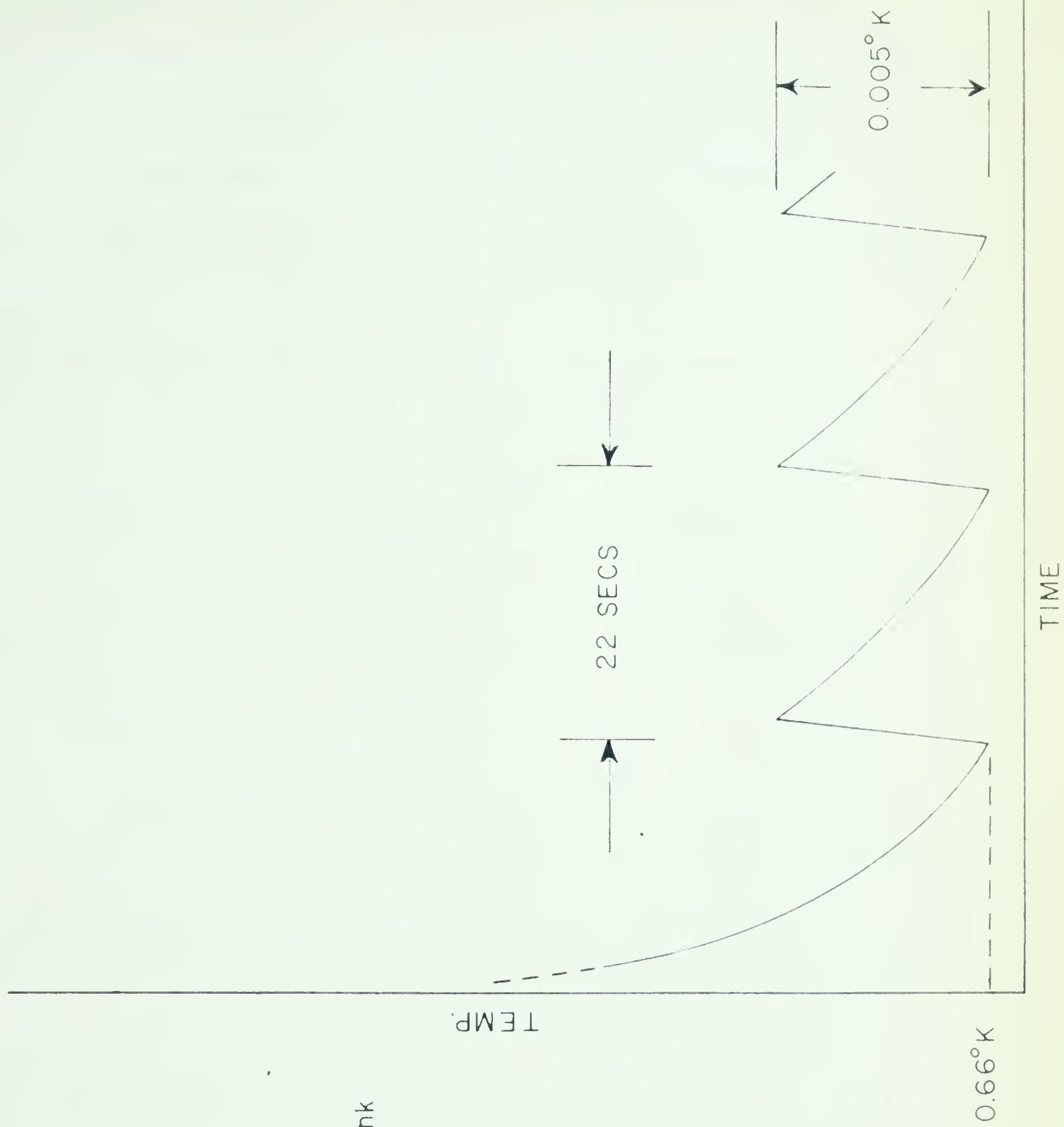


Fig. 1



of B rises because the film releases its heat of adsorption as it flows onto the surface of B. The heat of adsorption is of sufficient magnitude to account for the heat transferred in the process, while the thermo-mechanical effect is not. Almost immediately after the flow begins, it will be choked off because the temperature of B will have exceeded the onset temperature. The cycle will repeat as B cools again through the metallic link, C. Such a situation will give rise to relaxation oscillations, in which the period is essentially determined by the time in which the heat drains from the warmer body through the metallic link. Oscillations, typical of those actually observed in this laboratory, are shown in Fig. 1.

The static effect in which one body cooled when the other was heated must have been the result of the thermo-mechanical effect, causing the film to be desorbed from one surface, thus cooling it, and adsorbed on the other surface, thus warming it. In this work, emphasis has been placed on the temperature oscillations. The two phenomena are closely related with the difference that in the former, bodies of similar and rather large heat capacity are joined, while in the latter, the heat capacities of the two bodies are not comparable.

The first objective of this program has been to obtain temperature oscillations, for the typical experimental situation just described, under known and controllable conditions, and also to establish that the presence of an unsaturated helium film was necessary to the production of oscillations. Following this, the work has been directed toward testing the applicability of the proposed mechanism, as well as specifying certain qualitative features of the oscillations.

Chapter I deals with helium films, both saturated and unsaturated. The experimental apparatus is described in Chapter II, and the details of experimental procedure are covered in Chapter III. The experimental results, together with the interpretation of the temperature oscillation mechanism in the light of these findings, are given in Chapter IV. Chapter V is devoted to a discussion of the implications of this experiment with regard to properties of the unsaturated helium film.

CHAPTER I

HELIUM FILMS - SATURATED AND UNSATURATED

The main feature of the low temperature investigation to be described here is the helium film. This chapter will be used to introduce the subject of helium films generally, with particular emphasis being placed on the parts of the subject which are of principal importance in the present work. To begin with, it seems necessary to give a brief introductory discussion of some properties of liquid helium. The subject is treated in detail in the review by Daunt and Smith (1954).

Liquid helium boils at approximately 4.2°K , and at a temperature of about 2.2°K undergoes a liquid-liquid phase transition marked by sharp anomalies in the specific heat, dielectric constant etc. Below this transition temperature, usually referred to as the lambda point, the unique properties of the so-called Helium II phase appear. From a phenomenological viewpoint, the two-fluid model of Helium II (summarized in detail by Daunt and Smith) has proved to be most useful in interpretation of a wide range of experimental findings. The liquid is considered to be an intimate mixture of a "normal" fluid, density ρ_n , and a "superfluid", density ρ_s , such that $\rho_n + \rho_s = \rho$, where ρ

is the total density. The normal fluid is similar in behavior to Helium I (i.e. liquid helium above 2.2°K), while the superfluid has zero entropy, zero viscosity, and a total energy identical with the zero point energy of liquid helium. The relative proportion of these two fluids is strongly temperature dependent so that at temperatures as low as 1°K , very little normal fluid is present. At absolute zero, $\rho_n/\rho = 0$, while at the lambda point, $\rho_s/\rho = 0$.

One of the most important manifestations of superfluid flow is the thermo-mechanical effect (Allen and Jones, 1938). It is observed experimentally that when heat is introduced into part of a liquid helium bath, there is a flow of liquid in the opposite direction to the flow of heat, even when the path available for the flow is extremely narrow. In a typical experimental arrangement where a Helium II bath at temperature T is connected to a thermally insulated container of He II at a temperature $T + \Delta T$, a pressure gradient ΔP is induced by this temperature gradient. The magnitude of the effect is given by the London equation (London, 1939), viz $\Delta P = \rho S \Delta T$, where S is the entropy of the liquid at temperature T . In terms of the two-fluid model, this phenomenon may be viewed as superfluid flowing toward the

region at $T + \Delta T$, thus increasing the local concentration of superfluid, which in turn tends to reduce the temperature toward T . This is known as the thermo-mechanical effect. In the inverse mechano-caloric effect (Daunt and Mendelssohn, 1939), a temperature gradient is induced by a pressure gradient.

The nature of ideal superfluidity has usually been studied with the aid of very narrow flow channels, of the order of 10^{-5} cm. in width. The superfluid component of He II flows freely through such channels, in contrast to the normal component which is effectively immobilised by this barrier which prevents flow of the normal fluid because it has a finite viscosity. In addition to a variety of specially constructed flow channels (e.g. the gap between two optically flat pieces of glass), there is a "natural" flow channel of suitable dimensions - the helium film.

The Saturated Film

Any surface in contact with the bulk liquid or with its saturated vapor pressure is covered by a thick layer of liquid helium, referred to as the saturated film. For a detailed review of properties of the saturated helium film, see Jackson and Grimes (1958). Only a brief summary will be given here.

The helium film, first observed by Rollin (1936), is estimated to have an average thickness of the order of 3×10^{-6} cm., i.e. a number of investigations show that the film is 50 to 150 atomic layers in thickness. The saturated film possesses essentially the same static properties as the bulk liquid. If two volumes of He II, connected only by a film, have a temperature or pressure gradient between them, it is through the film that the thermo-mechanical or mechano-caloric effects occur. In the case of the saturated film, flow rate is almost independent of pressure head or length of flow channel, while temperature dependence is such that transfer rate falls rapidly to zero near the lambda point.

Most theories (e.g. Frenkel, 1940; Schiff, 1941) concerning the formation of the helium film involve the van der Waals forces between atoms in the film, and between helium atoms and the substrate. Additional effects due to zero-point energy have also been taken into account (Atkins, 1954; Franchetti, 1956, 1957). Each of these theories leads to an expression predicting film profile (variation of thickness with height above the liquid). Calculated film thicknesses on a barium-stearate coated wall (Franchetti, 1957) are larger than observed thicknesses, but are in good general agreement

with experimental measurements. In view of this agreement, the film may be regarded as adsorbed on the surface with which it is in contact. Further details on adsorption due to van der Waals forces will be given in the next section.

Helium films formed by the action of van der Waals forces should not, according to present theory, show any discontinuity in thickness upon passing through the lambda temperature provided no appreciable heat influx is permitted. Such thick He I films have been observed (Ham and Jackson, 1957) and were found to be extremely sensitive to extraneous radiation. The reason thick He I films are not usually observed is that liquid leaving the films cannot be replenished by superfluid flow from the bulk liquid. A similar situation may be found below the lambda temperature if the adsorption substrate is in contact with helium vapor alone, and cannot, therefore, be replenished from the bulk liquid. Bowers (1953) estimated that films no thicker than 40 or 50 atomic layers were obtained when a metal foil was suspended freely above a He II bath, while Kistemaker (1947) observed a thickness of about 150 atomic layers on a surface in contact with the liquid.

In contrast to the saturated film, a film may be formed, by van der Waals forces, in a system where the pressure of helium gas present is less than the saturated vapor pressure corresponding to that temperature. Such a film is referred to as unsaturated and it is the unsaturated film with which the experiment to be described here is concerned.

The Unsaturated Helium Film and Helium Adsorption

To say that a film in equilibrium with the vapor pressure of the surrounding gas, formed by van der Waals forces, exists on a surface is synonymous with saying that the film is adsorbed on that surface. Therefore an investigation of unsaturated film properties leads to the subject of gas adsorption. Although a considerable amount of work has been done in this field, it is only relatively recently that such studies have involved helium adsorption at low temperatures.

The process of adsorption is customarily described in terms of the adsorption isotherm, i.e. a plot of amount of gas adsorbed, v , (volume of gas per unit surface area) against relative saturation, p/p_0 , (p_0 is the saturated vapor pressure for a particular temperature). A more detailed discussion of properties

of adsorption isotherms, as well as a diagram of their characteristic shape is given in the review by Long and Meyer (1953).

Adsorption isotherms have been measured on a variety of adsorption substrates, for instance - Keesom and Schweers (1941a), on glass, and, (1941b), on solidified gas layers; Schaeffer, Smith, and Wendell (1949), on carbon; Mastrangelo and Aston (1951), on TiO_2 ; Strauss (1952), on Fe_2O_3 ; Bowers (1953), on aluminum foil; Meyer (1956), on gold foil. All adsorption isotherms have been determined by pressure measurement on a system of known volume, into which a known amount of helium had been admitted, with the exception of Bowers (1953) who measured the amount adsorbed directly by weighing with a special microbalance. Since the van der Waals forces differ for each surface on which adsorption occurs, certain quantitative disagreements in these isotherm measurements would be expected. However, data obtained about certain aspects of these isotherms are so divergent that it is impossible to state with any certainty whether or not, for a given relative saturation, amount adsorbed increases (Strauss, 1953; Bowers, 1953), decreases (Brewer and Mendelssohn, 1953), or remains constant (Tjerkstra, Hooftman, and von der Meydenberg, 1953), with decreasing temperature.

The heat of adsorption, or adsorption energy (i.e. the decrease in energy, per mole, for an infinitesimal quantity of gas adsorbed at a constant pressure) for helium is small in comparison to other gases. The reason is that part of the energy released by interaction with the wall is used to overcome the repulsion between atoms due to the zero point energy. Because of the van der Waals forces, energy of adsorption should decrease with the third power of distance from the wall, and be essentially independent of temperature. Keesom and Schweers (1941a) found adsorption energies independent of temperature, strongly dependent on degree of occupation. At an estimated coverage of 10% of the first layer, heat of adsorption was 100 cal/mole, decreasing to 40 cal/mole when approaching completion of a monolayer. Heat of adsorption on Fe_2O_3 (Strauss, 1952) for layers adsorbed at some distance from the surface was less than 25 cal/mole at 1.8°K , only slightly greater than the heat of vaporization of the bulk liquid. Measurements of adsorption energies have not been made at temperatures below or near to 1°K , the region of particular interest for the experiment described in this thesis.

The most widely applied theory of adsorption is that due to Brunauer, Emmet, and Teller (1938) - the BET theory. Helium adsorption data, when analyzed according

to the BET theory, indicate an anomalously high density in the first adsorbed layer. The quantity v_m , the volume of gas (at STP) adsorbed in the first layer, was evaluated at 3.6 times as large as the amount that would be contained in a monolayer of the density of the bulk liquid (Schaeffer, Smith and Wendell, 1949). This seemed reasonable in view of the fact that the volume of liquid helium is about four times that for a liquid of close-packed spheres having the gas kinetic diameter of about 2 \AA^0 , which is the result of the repulsive forces causing the high zero point energy of the liquid. However, a more thorough examination of the experimental results showed that the BET theory in its original form was not applicable at low saturations (Steele, 1956; Meyer, 1956) and disagreement with experiment had been generally observed for saturations above about 50% (Daunt and Smith, 1954). Bowers (1953) found that his experiments agreed well at high saturations with the Frenkel (1946) isotherm.

Aston and Mastrangelo (1951) have derived a modified form of the BET theory which proposes that the amount of helium v_m is contained in a solid multilayer film. Another theory of multilayer adsorption (Steele, 1956) predicts a first layer more dense than for bulk liquid packing, more dense than a solid layer,

about 40% less dense than estimates calculated from the BET theory. Steele quotes the calculations of Singh and Band (1955) which show that densities of the order of four times liquid packing lead to helium-helium repulsion energies so large as to completely overbalance any van der Waals attractive force. This theory shows satisfactory agreement with the adsorption data of Aston, Mastrangelo, and Tykodi (1951). Since it is probable that the quantity v_m is not the amount adsorbed in the first layer, Meyer (1956) suggests another interpretation in which v_m may be considered as the amount adsorbed under the condition $(E_s - E_o) > RT$, (E_s being the differential energy of adsorption, E_o the energy of vaporization of the bulk liquid). In the case of helium, this condition does not coincide with the completion of only one adsorbed layer.

The reason for the failure of the BET theory in achieving agreement with adsorption experiments may well be that the problem was oversimplified by one of the basic assumptions of the theory - i.e. only the first adsorbed layer has an adsorption energy greater than that of the heat of vaporization of the liquid. Steele considered the less restricted case in which the first two layers have adsorption energies different from each other and different from the heat of vaporization.

Up to the present time, no treatment has been given involving lateral interaction of adsorbed atoms, nor has the application of Bose-Einstein statistics accomplished any satisfactory agreement with experiment.

Most adsorption experiments have been performed prior to the latest theoretical work, cited above, and interpretation of part of the data has been based on the assumption that the first layer density was about four times that of the bulk liquid. The saturation at which the first adsorbed layer is formed is not well known, but it is almost certainly less than usually supposed, with the result that most experiments have probably been carried out when the first adsorbed layer was already present, i.e. it was the effective adsorbent. Bowers (1953), for instance, assumed that for $p/p_0 = 0.15$, just over one layer was present. Meyer (1956) suggests that what Strauss (1952) regards as completion of the first layer actually corresponds to completion of the second layer. Meyer's results indicate that the first layer, at 2°K , is complete for p/p_0 approximately 10^{-7} , while Hobson's (1959) measurements of helium adsorption on glass at 4.2°K , suggest that the first layer is complete at p/p_0 less than 10^{-12} .

In view of the findings reviewed in this chapter, it is often difficult to make any quantitative interpretation of experimental results given purely in terms of coverage according to statistical layers. Thus it would be easier to correlate data from different sources if it were presented in terms of amount of gas adsorbed per unit area, until some more satisfactory model is found.

Superfluidity in the Unsaturated Film

Like the saturated film, the unsaturated film is formed by van der Waals forces and may be several atomic layers thick. Since there are similarities between these films, one would expect that the behavior of the unsaturated film would in some way resemble the superfluidity found in the He II phase of the saturated film. In order to determine if, and under what conditions, the unsaturated film undergoes a lambda transition, three main aspects of the film properties have been studied - specific heat, film flow and heat transport.

Specific heat curves for helium adsorbed on Fe_2O_3 powder (jeweller's rouge) have been measured by Frederikse (1949, 1950) at coverages obtained between 6% and 80% of saturated vapor pressure. For a film thickness greater than about 20 layers, specific heat

was the same as the bulk liquid specific heat, while at lower coverages the specific heat maximum appeared less pronounced and was positioned below 2.2°K . For the lowest coverage studied, the data obtained indicated no specific heat maximum.

Long and Meyer (1952a) have employed several techniques in the study of flow characteristics of the unsaturated film. In one set of measurements (their Method I), flow rate from an adsorbent (Fe_2O_3), through a superleak, into an initially evacuated space was determined, for several pressures which were less than the saturated vapor pressure for the prevailing temperature. It was found that for any given saturation in the adsorption chamber, superfluid flow appeared abruptly below a certain temperature. The results of this experiment are shown by the open circles in Fig. 2, which illustrates the dependence of onset temperature on relative saturation. These onset temperatures are as much as 0.4°K lower than the above-mentioned peak in the specific heat curves of Frederikse. Flow measurements in the system just described were not made directly but depended on the assumption that all the liquid helium which passed through the superleak into the volume originally at high vacuum would evaporate there, thus enabling a subsequent determination of flow depending

on the pressure measured. However, in their second set of experiments (Method II), superfluidity appeared at all temperatures below the normal lambda point. Cooling experiments involving a temperature gradient between two chambers of adsorbent (Fe_2O_3) at less than saturated vapor pressure were also done, with the result that the thermo-mechanical effect, i.e. film flow, appeared at all temperatures below the normal lambda temperature. The apparent inconsistency of this work has never been fully explained.

Inconsistent results have also been produced in studies of film flow through porous "vycor" glass. Because the flow channels found in "vycor" glass (mean pore diameter 5×10^{-7} cm.) are narrower than the thickness of the saturated film (about 10^{-6} cm.), this super-leak is of particular interest because it is of similar size to an unsaturated film. Champeney (1958) found no onset temperature lower than 2.07°K , while Atkins and Seki (1957) observed an onset temperature as low as 1.36°K .

Another approach to the investigation of unsaturated film flow is through experiments concerning heat transport. Bowers, Brewer, and Mendelssohn (1951) found that the unsaturated film was able to transport heat, with zero temperature gradient, between a heat sink and

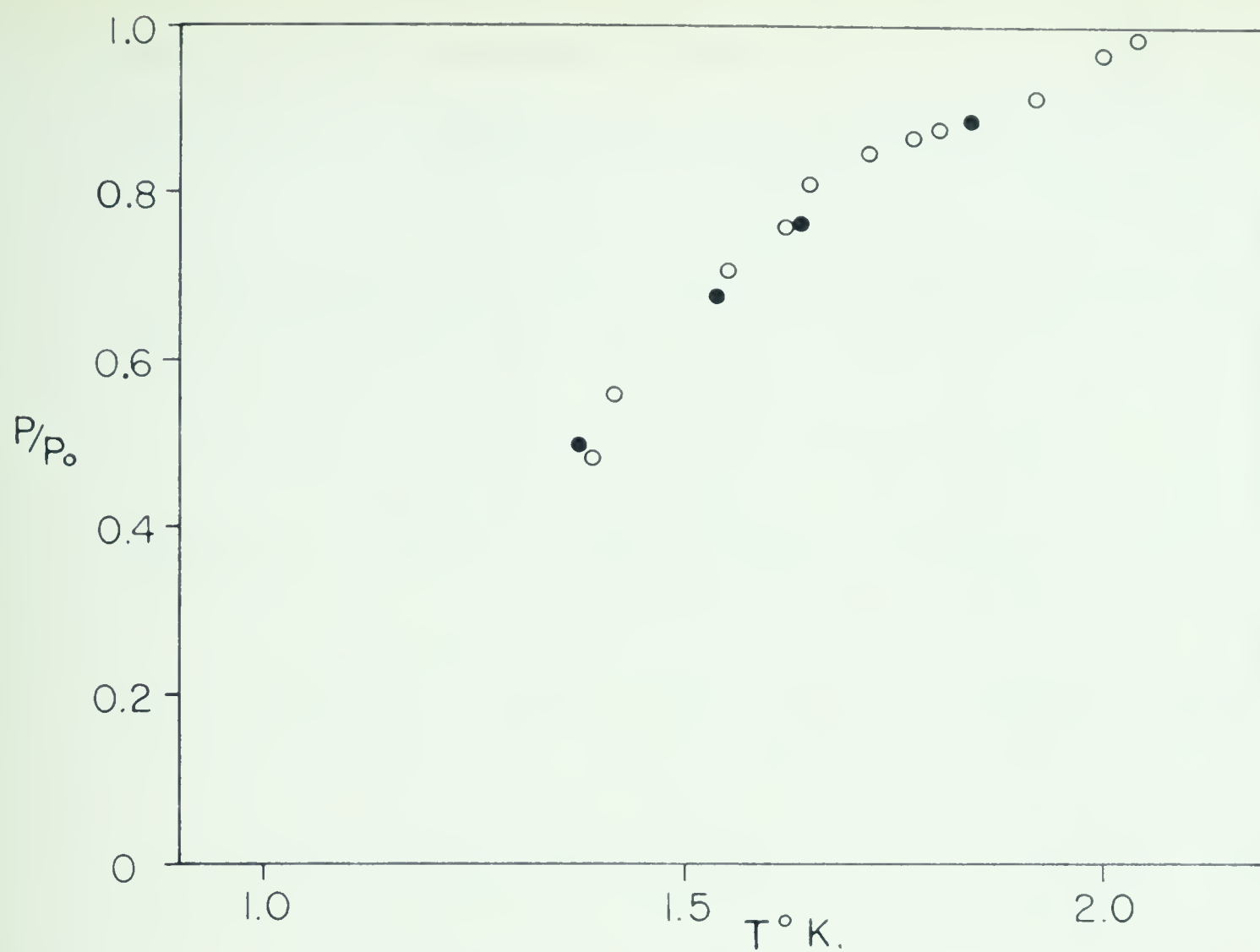


Fig. 2 - Onset of superfluidity as a function of relative saturation pressure

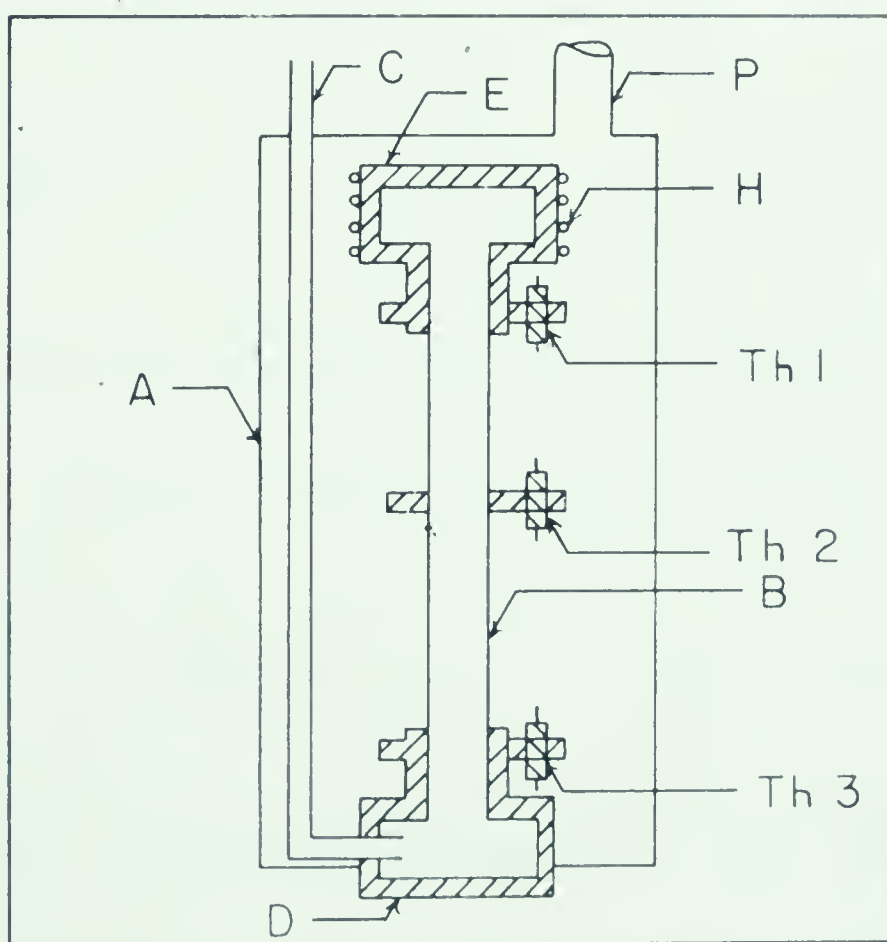


Fig. 3 - Heat transport apparatus used by Long and Meyer

a chamber connected to it by a thin-walled tube, and supplied with a heater. When a critical heating rate was reached for a given saturation, temperature of the top chamber rose in a "run-away" phenomenon. Temperatures corresponding to a critical heating rate of zero for each saturation are shown by the solid circles of Fig. 2.

Using a similar method, Long and Meyer (1952b, 1955) undertook a further study of heat transport in the unsaturated film. Their apparatus is shown schematically in Fig. 3. At the top of a copper-nickel alloy tube, B, there was a copper chamber, E, wound with a heater, H. At the bottom of B, there was another copper chamber, D, immersed in the He II bath which surrounded the vacuum can A. E was essentially thermally isolated from D by the high vacuum in A, obtained through pumping line P, and also by B, a very poor thermal conductor. Carbon thermometers, Th 1, Th 2, Th 3, were placed at the top, middle and bottom of B. Helium gas was admitted to these chambers through the capillary tube C.

The onset temperatures found by Long and Meyer were consistent with those of Bowers et al, but they found that heat transport occurred with a finite temperature gradient between E and D, which increased non-linearly with increasing heat current. The temperature

distribution along the tube B led them to conclude that a heat transport cycle must take place, i.e. superfluid film flows to some higher temperature region, evaporates there, and returns as gas to the heat sink, D. Condensation occurs at D, and the cycle repeats. They also suggest that, at a given temperature, a certain number of layers of film are immobile, so that superflow only occurs if more than this number of layers are present in the film.

White, Chou and Johnson (1952) have reported that superfluid contribution to heat transport was not observed except at very high saturations, in an experimental apparatus similar to that of Fig. 3 but the tube equivalent to B was packed with silica gel, which would inhibit gas flow in a heat transport cycle.

A recent theoretical study (Hecht, 1958) employing a partition function derived by Feynman, deals with lambda points in unsaturated helium films. After a reassignment of film layers, Hecht's equation successfully predicted the temperatures of the peak in the specific heat curves measured by Frederikse (1949). He suggested that the second film layer, the first liquid layer, had usually been assigned too great a density (i.e. half the density of the first layer, assumed to be four times the density of the bulk liquid. This is

discussed earlier in this chapter.) and added $\frac{3}{4}$ of a layer to the experimental data to which the theory was applied. The first layer was assumed to be solid and did not figure in the theory. From the point of view of the present work, the most interesting part of Hecht's theory is the prediction that a transition to superfluidity will first be found in a film not thinner than $1\frac{1}{2}$ layers, at a temperature of 1°K . This theory does not agree with the experimental values of temperatures for onset of superflow, but a reason is suggested why the heat capacity peak and the onset of superfluidity should occur at different temperatures. If some of the top layers in the film were only partially filled, the average number of nearest neighbors to any given atom would be reduced, thus reducing an onset temperature which would depend on a cooperative effect of all the atoms, but not affecting the heat capacity which would still depend on average thickness.

It is evident that behavior of the unsaturated helium film is not yet completely known, but certainly some typical He II properties appear in these films under experimental conditions of the sort just described. The review article by Long and Meyer (1953) points out several questions about unsaturated films not answered adequately by experiments up to that date, and which still have not been satisfactorily investigated at the present

time. Little is known about the possibility of a sharp transition point in the unsaturated film corresponding to the sharp definition of the lambda point in the bulk liquid. The importance of a heat transport cycle in superfluid flow is not yet clear. Rate of evaporation in such a heat transport cycle, and not mobility of the film, might be the rate-determining factor in phenomena involving unsaturated film flow. The two-fluid model may not be applicable to some observed properties of these films. Such questions as these can only be resolved by new experiments. It is possible that the experiment described in this thesis, aside from explaining the unusual thermal behavior described in the introduction may be of some use in understanding the nature of the unsaturated helium film.

CHAPTER II

EXPERIMENTAL APPROACH AND THE APPARATUS

An experiment designed to detect temperature oscillations of the type described in the introduction required an adiabatic demagnetization cryostat containing an experimental chamber having the features shown in the inset of Fig. 1. This chapter will be used to describe the apparatus used to perform the experiment, beginning with the details of that part of the equipment necessarily original to this experiment, namely the experimental chamber and the helium gas filling system. This will be followed by a general description of the cryostat, typical of many used for demagnetization experiments, as well as the electrical instruments and the pressure measuring devices, from which the experimental data has been obtained.

The Experimental Chamber

The general configuration of the experimental chamber has already been given in the inset of Fig. 1. It is shown again in Fig. 6 in its position relative to the whole low temperature end of the apparatus, and the construction details of it may be seen in the photograph of Fig. 4. In general outline, the experimental situation

may be stated as follows. The warm body, B, was required to have a small heat capacity in order to have good sensitivity to heat deposited by small amounts of helium flowing onto its surface, while the cold body, the paramagnetic salt surrounding A, was required to serve as a cold reservoir. The link, C, joining these bodies was chosen to have poor thermal conductivity in order to limit the thermal contact between A and B and thus maintain a temperature gradient.

The paramagnetic salt, and the tube A, were contained in the capsule D. (Refer to Fig. 6 for explanation of the lettering used in this section.) A copper tube, A (0.188" ID, 0.212" OD, 2.25" long), was situated at the center of D, projecting through an opening in the top of D. The tube A had a brass bottom, and a system of eight fins of thin copper foil projecting diametrically into the annular space between the copper tube and the outside of D, which was a section of inconel tubing (0.75" OD, wall thickness 0.010", length 2"). Top and bottom caps of D were made of brass, thickness about 0.03".

A small copper plate was soldered to the top of A just above the outside of D for the purpose of mounting a thermometer in a position where it could be in good



—— the helium
pot

—— the experimental
chamber

Fig. 4

thermal contact with the interior of D by means of an all-copper conduction path. The carbon thermometer, shown symbolically as T_2 , was a $\frac{1}{2}$ watt, nominal 10 ohm, Allen-Bradley resistor, prepared as follows. After the outside coating had been ground off, it was insulated with two coats of GE Baking Varnish, baked on separately. The thermometer, T_2 , was then baked onto the copper plate using the same varnish. For characteristics of the carbon thermometers used, see Appendix I.

The capsule D was filled with powdered potassium chrome alum, $KCr(SO_4)_2 \cdot 12 H_2O$, mixed with glycerine in the proportion 52.0% to 48.0%⁺. Total weight of the paramagnetic salt was 9.31 gms. The mixture of glycerine and salt was thought to be a good way of achieving thermal contact with the copper fin system. The bottom cap of D was soldered with Wood's metal in order to protect the salt, which has a decomposition temperature of about 60°C.

The thermal link, C, was made of inconel tubing (0.125" OD, wall thickness 0.004", length 1.34"), and was joined to the top of A and the bottom of B with brass ferrules. The canister B was made of copper tubing, cut to a length of 1.5", turned to a wall thickness of about 0.010", and 0.53" ID. Top and bottom caps were made of brass. Joints on A, B, C, and D were made with silver solder or soft solder (50% Pb - 50% Sn).

+ by weight

After an insulating coat of baking varnish was applied, a heater, H, was wound on the top half of B, and another layer of varnish was baked over the top of H. The heater was made of manganin wire, double silk-covered, B & S No. 44 gauge, resistance of 74.48 ohms per foot, total resistance of 460 ohms. A carbon thermometer, T_1 , was prepared in the same way as T_2 , and attached to the surface of B. A fine capillary tube, E, was used either as a pumping line or helium filling line for the interior of A, B and C, and will be described in more detail in connection with the helium gas filling system.

To supply variable thermal linkage between the paramagnetic salt capsule (hereafter referred to simply as the salt) and the insulated copper canister (hereafter referred to as the can), they were joined with a superconducting thermal switch, L. It is known that, by application of a small magnetic field, a superconductor may be restored to normal conductivity while at a temperature below its usual transition temperature. Thermal conductivity of a normal conductor is considerably greater than that of the same element at the same temperature in its superconducting state. Thus when two bodies such as A and B are joined by a superconducting link, such as L, a marked change in heat flow between the two

bodies is obtained by thermal switching, i.e. by applying the magnetic field required to cause a transition in the link. Detailed properties of superconductors are discussed in the book by Shoenberg (1951). The experimental data summarized by Heer, Barnes and Daunt (1954) shows that, at 1°K , thermal conductivity of lead varies by a factor of about 60 upon undergoing a transition from the superconducting state to the normally conducting state. The link, L, shown in the photograph of Fig. 4 was a wire about 12" long, diameter 0.014". A rough calculation shows that this amount of lead gives thermal linkage between A and B of the same order of magnitude as that provided by the inconel tube C. At 1°K , a magnetic field of about 800 gauss is required to operate the thermal switch. Purity of the lead used in L was not known, but later in a modified design (to be described in the next section), lead of 99.999% purity was used.

The whole of the apparatus just described was suspended by nylon threads in the cradle, K. The cradle was a cylindrical brass framework which could be screwed onto the bottom of V_3 . The nylon thread assured a firm suspension without thermal contact with the cradle, since the thread has a very low thermal

conductivity of 0.06 milliwatts/ deg. - cm. (Berman, Foster, and Rosenberg, 1954).

Electrical leads for the thermometers and heater entered the experimental space through a platinum-glass seal, G, which had the required eight leads plus two extras. Although a certain amount of heat leak from G to A and B due to the introduction of the electrical leads could not be avoided, they were designed to minimize joule heating due to the thermometer and heater currents. To prevent heat leakage due to thermal conduction, it was necessary to use wire of small diameter, of a length as great as the available space permitted. Each lead was a two or three foot length of manganin wire, B & S No. 40 gauge. Ideally, the wires should be superconducting to prevent joule heating, but the manganin wire had a high resistivity. However it had a much lower thermal conductivity than copper wire, for instance. Wire fitting the above requirements is not available commercially, so the manganin wires were tinned with 50% Pb - 50% Sn solder, which would be superconducting at low temperatures. The wires were insulated with three coats of GE Baking Varnish, baked on by electrical heating of the wire. Wire insulated in this way had an uneven appearance, but no electrical shorts due to insulation failure were experienced. For orientation of the leads, see Fig. 4.

Modifications of the Experimental Capsules

To test the influence of geometry on the temperature oscillation phenomena, several different tubes were placed in position C. The design just described in detail will be referred to as arrangement 1, and it may be distinguished by noting that the tube C had a diameter of 0.125".

Because of the method of assembling arrangement 1, it was necessary to construct another copper canister, and another salt capsule, both identical with those already described, except that the carbon thermometer on the salt capsule was encased in a small rectangular copper block with a cylindrical hole in it. Although it was still necessary to use several coats of baking varnish as insulation, it was hoped that better thermal contact between salt and thermometer would be achieved in this way. The new salt capsule contained 13.0 gms. of paramagnetic salt, more than the previous capsule. A special set of joining ferrules was made so that a variety of metal tubes could be placed in position C without damage to the rest of the assembly.

For the modification known as arrangement 2, a small diameter (0.033" ID, 0.040" OD) cupro-nickel alloy tube was used, of the same length as C in the

original design, arrangement 1. For arrangement 3, the length of C was also the same as previously used, but it had a larger diameter (0.247" OD, 0.231" ID) and was made of stainless steel. The thermal switch, L, was the same piece of lead wire for both of arrangements 1 and 2, but in arrangement 3, the link consisted of four 6" lengths of wire 0.020" in diameter, made of lead 99.999% pure.

The Helium Gas Filling System

Since the internal surfaces of the capsules described in the last section functioned as a helium adsorption chamber, it was necessary to have some method of measuring the amount of helium gas admitted to the helium adsorption chamber and of subsequently measuring the equilibrium pressure over the adsorbed helium layers. The apparatus to be described in this section is shown in Fig. 5.

Helium gas, admitted through stopcock A, was cleaned in a liquid air cooled charcoal trap, isolated by C and D, and then admitted to a 500 ml. storage bulb, isolated by D and E. The desired amount of helium, as measured by the oil McLeod gauge and the oil manometer (to be described under Pressure Measurement), was held

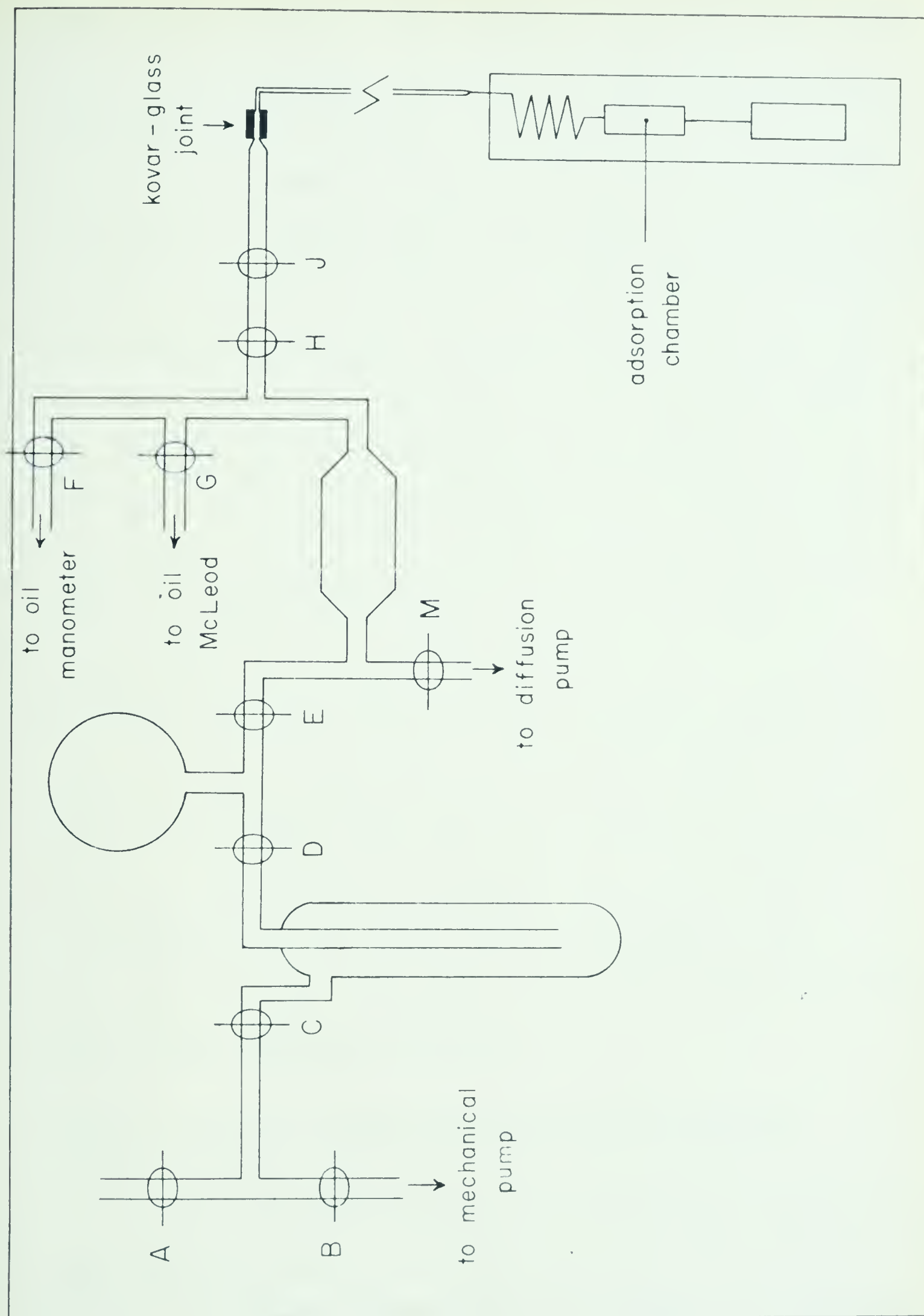


Fig. 5 - The helium gas filling system

in the volume enclosed by E, M, F, G, and J and was admitted through J to the adsorption chamber via a long metal capillary tube. The small volume between H and J was found to be 0.930 cc. Several other volumes in the glass filling system were calibrated simply by filling the 0.930 cc. volume with a known amount of gas, expanding it into a previously evacuated volume, and then measuring the resulting pressure. The cupro-nickel filling tube was about three feet in length, 0.031" ID, and entered the pumping line for the experimental chamber at the radiation trap, as shown in Fig. 6. At the top of the experimental space it was joined to a finer capillary, made of cupro-nickel alloy, (0.020" OD, 0.008" ID, length about 30"), and it is denoted as E on Fig. 6. By using this long section of fine capillary with low thermal conductivity, most of the heat leak to the adsorption chamber from that part of the filling line which passed through the helium bath at 4.2°K was eliminated, but even with these precautions, this capillary tube, E, was probably the major source of heat leak to the adsorption chamber.

General Features of the Demagnetization Cryostat

The cryostat used for this work was typical of many cryostats used to do experiments in the liquid helium temperature region. It was similar, for instance,

to an apparatus described by White (1955) for use in experiments involving adiabatic demagnetization. Very generally, the design of such an apparatus is aimed at providing an experimental space, refrigerated by liquid helium, which may be thermally isolated from associated parts of the apparatus by a high vacuum, or which may be put in thermal contact with a cold reservoir which must be capable of operation at several different temperatures.

The low temperature section of the equipment, shown in Fig. 6, was immersed in a liquid helium dewar which in turn was immersed in a liquid air dewar. Both dewars were made with narrow tails in order to be accommodated by the 3" pole gap of the magnet (to be described later). The liquid helium dewar had an overall inside length of 35", inside diameter 85 mm. at the top, and inside diameter 40 mm. in the tail. The liquid air dewar had an inside length of 30", top inside diameter 130 mm., tail inside diameter 60 mm.

Liquid helium used in this experiment was produced by the Collins liquefier in the Physics Department of the University of Alberta. All helium gas produced as the liquid boiled away was automatically fed into a gas recovery line, Z, and later reliquefied. The helium dewar was covered by a dewar cap, Q, suspended

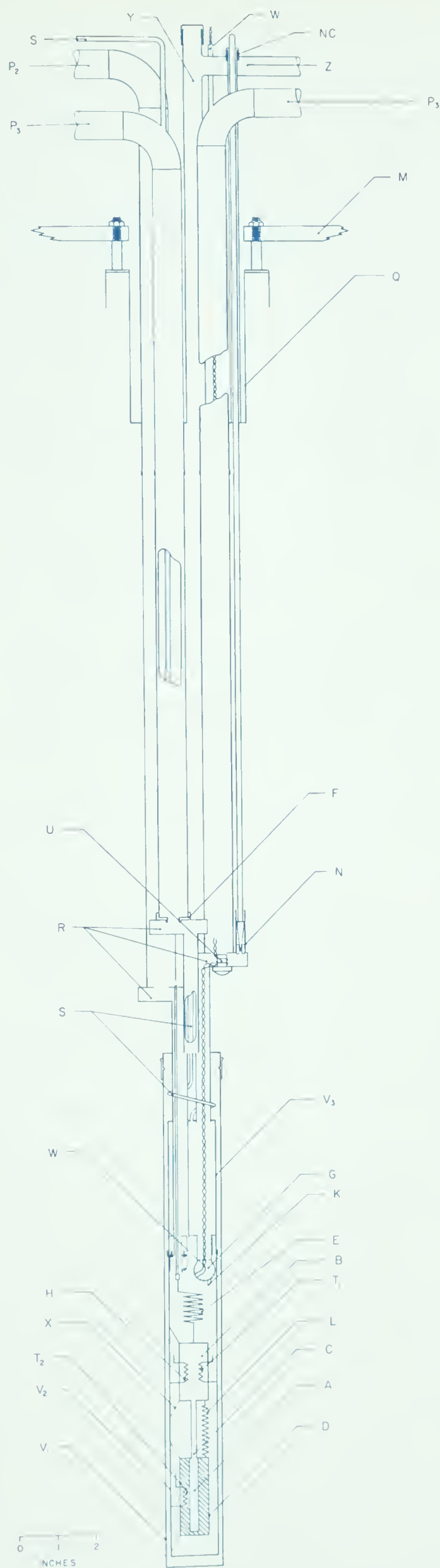


Fig. 6

from an aluminum plate, M, which was attached to horizontal arms projecting from a "Hammond" relay rack. This dewar was joined to the cap by rubber tape, and liquid was transferred to it from a liquid helium storage vessel by a double-walled syphon, entering the dewar through Y.

The vacuum jacket, V_2 , enclosing the experimental space was in turn enclosed by another vacuum jacket, V_1 , which was used to isolate the contents of V_2 from the helium bath at 4.2°K . The chamber V_3 will be described shortly. V_1 and V_2 were brass cylinders, wall thickness about $1/32"$. The three pumping lines, P_1 , P_2 , and P_3 , were made of $\frac{3}{4}"$ diameter thin-walled inconel tubing, which was intended to minimize heat leakage due to thermal conduction from the room temperature end of the tubes. To eliminate radiation down the pumping tubes from the room temperature end, the pumping lines were broken by insertion of small rectangular brass boxes, R, which functioned as radiation traps. Pumping lines below the traps R were of $\frac{3}{8}"$ inconel tubing. Joints between these metal pieces, connected by brass ferrules, F, were made with soft solder or silver solder, but V_1 and V_2 could be easily removed because they were joined with Wood's metal.

The reservoir, V_3 , known as the "helium pot" was filled with liquid helium by opening the needle valve, N, operated at room temperature by raising or lowering it in the needle valve casing, NC. The helium pot, made of copper to aid in even heat distribution, served as a heat sink for the experimental chamber. Its temperature could be varied from 4.2°K to a minimum of 1.2°K , by reducing the vapor pressure over the boiling liquid helium, which was pumped through the line P_3 . Vapor pressure in the helium pot was measured by a mercury manometer and an oil manometer (to be described under Pressure Measurement) connected to V_3 by S, made of $\frac{1}{8}$ " diameter inconel tubing.

Low vapor pressure in the helium pot was provided by a mechanical pump with a speed of 100 liters per minute. V_1 and V_2 were evacuated by liquid air trapped mercury diffusion pumps, backed by mechanical pumps. The pump for the line P_2 was also used to evacuate the helium gas filling system and adsorption chamber.

Thermal equilibrium between two bodies normally in poor thermal contact is usually produced by surrounding them both with a low pressure of helium "exchange gas". A glass exchange gas storage system was mounted on the

Hammond rack. Helium gas could be admitted to either V_1 or V_2 , which could be isolated from their respective pumps.

The magnet used in this experiment, made by Pacific Electric Motor Co., Model 12A-H1, was capable of supplying a field up to 11,500 gauss with a 3" pole gap. A roll of copper foil was placed inside the liquid air dewar to impede possible eddy current heating in all copper parts of the apparatus because of "ripple" in the current provided by the magnet power supply. Several limitations in the cryostat design were imposed by the fact that the widest available pole gap was 3", at a fixed height from the floor. In addition, the magnet was permanently fixed in one location, so it was necessary to place the experimental chamber in the pole gap by moving the whole cryostat forward on rails mounted on a thick plywood board.

Pressure Measurements

High vacua in V_1 , V_2 , and the helium gas filling system were measured by a Philips type cold cathode ionization gauge (made by H. S. Martin and Co.). Typical vacua would be as follows - at room temperature, 2×10^{-6} mm. Hg; at liquid air temperatures, 1×10^{-6} mm. Hg; at liquid helium temperatures, 6×10^{-7} mm. Hg.

A gas discharge tube was used to make a qualitative estimate of exchange gas pressure before filling jackets V_1 or V_2 .

Vapor pressure in the helium pot was determined by the use of two manometers, mounted on a board at one side of the Hammond rack, evacuated continuously on the reference side. These vapor pressure readings were converted to absolute temperatures using the tables compiled by Clement (1956), for the "1955E" temperature scale as defined by Clement, Logan and Gaffney (1955). Pressure control could be obtained when necessary, such as during thermometer calibration when the thermometers were in exchange gas contact with the helium pot, with a Cartesian manostat connected to the helium pot pumping line, as described by White (1959).

Pressure in the helium gas filling system was measured by an oil manometer and another device, similar in principle to the conventional mercury filled McLeod gauge. This gauge, employing low vapor pressure oil as the working substance, was first used by Nester (1957), and will hereafter be referred to as the oil McLeod gauge. It is described in detail in Appendix II. With these two gauges, the amount of helium input to the adsorption chamber could be measured, and the room temperature value

of helium gas pressure over the unsaturated film formed there could also be measured. According to evidence obtained in previous experiments (see Chapter I), an unsaturated helium film having a transition temperature near to 1°K is very thin, and in equilibrium with a very low vapor pressure of helium gas. Before oscillations were actually observed, a Bayard-Alpert gauge, and a high vacuum McLeod gauge were connected to the helium filling system to measure the expected low percentage of saturated vapor pressure, but these were later disconnected when it became evident that the range of measurement for these gauges was unsuited to the actual conditions observed during the temperature oscillation investigation.

Electrical Measurements

Electrical leads, W, from the seal, G, were necessarily a source of heat influx to the helium pot, but it was found that the time occupied by one filling of the helium pot could be improved from $\frac{1}{2}$ hr. to $10\frac{1}{2}$ hrs. by a suitable choice of wire, thermally anchored at 4.2°K . Each lead was a one meter length of B & S No. 44 gauge, double formvar insulated copper wire, resulting in a heat influx through each lead of about 6000 ergs/ sec., by thermal conduction. Although manganin wires have a much lower thermal conductivity, joule heating due to current

flowing in the wires would cause a similar heat influx. Most of this heat was dissipated by thermally anchoring the leads at 4.2°K , i.e. winding the leads around an insulated copper plug, U. The formvar insulation of this wire proved inadequate for these conditions of close confinement, so it was necessary to separate the wires by spacing them singly on the sticky side of a long thin strip of masking tape. These fine wires were connected to the wires leading to the external measuring equipment at an all-copper terminal strip located at the top of the plate M.

Current for the thermometers was supplied by a simple circuit consisting of a battery in series with a continuously adjustable high resistance and the two thermometers. Current values up to 20 microamperes could be obtained and were measured by determining the potential drop across a 1000 ohm standard resistance.

Current to energize the can heater was drawn from a circuit similar to that of the thermometer supply, with maximum heater current being one milliamperere. A dummy heater, having approximately the same resistance as the heater and leads, was used to allow pre-adjustment of the current which was to be supplied to the can heater.

The potential drop across each of the carbon thermometers was determined by balancing a vernier potentiometer (manufactured by Cambridge Instrument Co.) for zero galvanometer deflection. This instrument was capable of detecting potential differences down to 10^{-7} volt. However, except for thermometer calibration measurements, the potential across T_1 , the can thermometer was continuously measured on a recording potentiometer. All measurements for experimental arrangement 1 were obtained with a Fisher Recordall instrument, range 0 - 5.5 MV, while for arrangements 2 and 3, a Leeds and Northrup Speedomax, Type G Recorder, Model S, 60000 Series was used. A bucking voltage was supplied in opposition to the thermometer EMF so that the zero of chart readings could be adjusted to a value just below the relatively high potential drop across T_1 , and the voltage oscillations could be displayed over nearly the full chart width, with a recorder range of 0 - 5 MV.

CHAPTER III

EXPERIMENTAL PROCEDURE

Preparation for an Experiment

Before beginning an experimental run, it was necessary to ensure good thermal insulation by thorough evacuation of vacuum spaces V_1 and V_2 (of Fig. 6), preferably by pumping overnight. Care was required in cooling the apparatus, because of the danger of placing undue strain on the platinum glass seal if it was cooled too rapidly. First the parts of the apparatus outside jacket V_1 were cooled by immersion in a dewar of liquid air. In a few minutes V_1 was filled with helium exchange gas, and later when the outside of V_2 was cooled, the inside of V_2 was filled with exchange gas, thus cooling the contents of the experimental space to liquid air temperatures.

At this point it was possible to begin the transfer of liquid helium into the empty inner dewar, which was sealed and connected to the automatic helium gas recovery system. The first part of the helium liquid transferred from the storage vessel to the dewar through the double-walled syphon was turned to gas during the process of cooling the apparatus to liquid helium temperatures. When enough liquid had accumulated, the helium

pot, V_3 , was filled by opening the needle valve. The helium pot was pumped to lower temperatures after breaking thermal contact between it and the helium bath at 4.2°K (in the helium dewar) by pumping away the exchange gas in V_1 . Usually the temperature of the helium pot fell to 1.2°K in about twenty minutes.

Cooling by Adiabatic Demagnetization

It was usually just before magnetization that helium gas was measured and admitted to the adsorption chamber. The heat produced by adsorption of this warm gas on the can could be seen on the chart recording, the warming being especially prominent if no exchange gas was present in V_2 , the experimental chamber.

The thermodynamic principles involved in the process of adiabatic demagnetization are well known and many experiments have been performed since the first adiabatic demagnetization produced by Giauque and MacDougall in 1933 (see Zemansky, 1957).

The procedure followed in the work outlined here was the same as that described in detail in a paper by Hull (1947) on the technique of adiabatic demagnetization, with the important exception that the method of temperature measurement was different. The temperature of the paramagnetic salt is usually determined by measuring its

magnetic susceptibility, where the salt sample is the core of a self or mutual inductance. The susceptibility gives a measure of temperature on an arbitrary scale, by an extrapolation of Curie's law. This temperature is then converted to the absolute scale of temperature as described by Hull. It was not possible to follow this procedure for measuring temperature in this experiment since it would have necessitated separation of the magnet and the cryostat. Since the magnet was fixed in one location, the cryostat would have had to be moved, which would have caused a serious disturbance in the experimental chamber due to heating by vibration. In any case, another type of thermometer was required for the copper canister.

Before beginning magnetization of the salt capsule, the experimental space was filled with helium exchange gas, thus providing thermal contact between the experimental samples and the helium pot, at its minimum temperature, 1.2°K . Usually magnetization of the salt to full field of 11,500 gauss was accomplished in about four steps. The heat of magnetization distributed throughout the salt and can was gradually removed by the exchange gas contact with the cold reservoir. The record of can temperature as the field was increased was always observed as a rapid temperature rise (typically

about 0.5°K) followed by a slow cooling to the helium pot temperature. The general appearance of this warming and cooling of the can is shown at the right of Fig. 10, which has been traced directly from the chart record. The dotted line indicates the way in which cooling occurs when no helium gas is present in the adsorption chamber, and the solid line indicates the thermal behavior of the can when an unsaturated film, which undergoes a lambda transition as it passes through that temperature range, is present there. The reason for this behavior will be explained in Chapter IV.

When the salt was magnetized to full field and the salt and can were in equilibrium at the helium pot temperature, the exchange gas was pumped from the space V_2 . Usually only five or ten minutes were required for the vacuum to reach about 2×10^{-6} mm. Hg, as read on the ionization gauge. The paramagnetic salt was then demagnetized adiabatically from full field to zero field (or some small non-zero field if observing the thermal switch effects) over a period of less than one minute. A sudden drop in the magnetic field would have resulted in unwanted eddy current heating in the can. The temperature drop of the can was followed on the chart recorder using a constant range of 0 - 5.5 MV, by adjusting the zero of the potential reading with the bucking

voltage applied to the input of the recorder. The salt temperature was followed on the potentiometer. Occasionally, balancing the potentiometer would cause a disturbance on the recording of the can potential, but this was noted when it occurred.

In designing the experimental chamber, it was necessary to position the lead wire between can and salt in the magnet just above the flat part of the pole pieces, thus placing it in a fringe field about 25% less than that between the flat part of the pole pieces. It was found that a superconducting transition was assured by changing the magnetic field from 1270 gauss to 660 gauss. For a "dry run", without an adsorbed helium film, the lowest temperature reached by the can after demagnetization could be considerably reduced by leaving the thermal switch on, i.e. normally conducting at 1270 gauss. If the final field after demagnetization was 660 gauss or less, a definite cooling of the can could always be observed by putting the thermal switch on. Whatever the means of thermal contact, a temperature difference always existed between A and B with the can being warmer by 0.2°K to 0.5°K . This may be attributed to the small natural heat leak to the can which must have exceeded the heat removed from the can by the link C.

The Observation of Temperature Oscillations due to
Unsaturated Film Flow

Temperature oscillations of the can, B, were found to occur spontaneously within one minute of adiabatic demagnetization of the salt, providing a sufficient quantity of helium gas was adsorbed on the inner surfaces of A, B, and C. The possibility that oscillations would begin was indicated by a much more rapid and pronounced cooling of the can than would be observed for a "dry run", for example T_1 would be 0.6°K , instead of 0.8°K . As they appeared on the chart, it could be seen that the oscillations were of the relaxation type - a rapid temperature rise would be followed by a slower, exponential cooling to the original temperature. The oscillations continued for five or ten minutes with a constant period of about twenty seconds. The amplitude of the oscillations was constant, except for the first one or two cycles, which were often difficult to observe because they passed through more than the full scale range of the chart recording. The shape of the oscillations was occasionally obscured by slight vibration of the apparatus or "noise" in the thermometer circuit. The effect of the magnetically operated thermal switch was sometimes observed by changing its state during a set of oscillations.

The end of a set of oscillations was indicated by perhaps one or two small amplitude cycles, and then both can and salt temperature rose rapidly at first, and then more slowly, to the region near 1°K , the salt always remaining cooler than the can.

The way in which a lambda transition in the film was observed is shown on the left side of Fig. 10. At some temperature above 1°K , the can heater was turned on. A pronounced change in warming rate of the can was noticed as the transition was passed. The heater was then turned off and the can cooled through the same transition, as marked in Fig. 10. These effects will be explained in Chapter IV.

The pressure in the helium gas filling system, i.e. the pressure in the adsorption chamber as observed at room temperature, was recorded at intervals during the whole procedure described above.

The number of demagnetizations, followed by a set of oscillations, that could be performed in, say, a twelve hour period, varied from four to eight, depending on conditions encountered during the course of the experiments.

A considerable amount of work was done using experimental arrangement 1, so that it is possible to form a fairly complete picture of its thermal behavior. However only a limited number of experiments, of a preliminary nature, have been done with arrangements 2 and 3, so only a general impression of their behavior has been obtained up to the present time. Experiments were interrupted by the necessity of dismantling all low temperature equipment in this laboratory and setting it up again in the new Physics Building. It is hoped to resume the temperature oscillation experiments in order to clarify some of the details of the unsaturated film behavior which should be known to complete this investigation.

CHAPTER IV

EXPERIMENTAL RESULTS

Temperature oscillations due to unsaturated film flow have been observed under the conditions described in Chapter III. On the basis of these observations, a mechanism for the oscillations has been deduced. The main features of it are substantially the same as the originally proposed mechanism (see Introduction), but with several important differences. According to the information available at the present time, the oscillations are thought to occur in the following manner.

When the salt around A (the notation refers to Figs. 1 and 6) is adiabatically demagnetized, the temperature of B drops due to conduction through the tube C, through the lead link, through the helium gas present in C, and by means of desorption of the helium film originally present on B. The helium gas released at the warm end of the system is "pumped" toward the cold end, and adsorbed there. No significant heating of the salt will occur since it has a large heat capacity. The film present on B is then extremely thin, and thus its transition temperature is much decreased from the value it had before demagnetization of the salt. Final

distribution of the film immediately after demagnetization depends on the temperature gradient between A and B, and on the amount of helium in the system.

Film flow toward the warmer end begins as soon as B is cooled through the transition temperature for the film thickness present there. The heat of adsorption is deposited on B, warming the can and the film until the film has passed its transition temperature, and the flow is stopped. The can is cooled once again by desorption of the recently deposited film, and in addition it is cooled by thermal conduction through the metallic link, which, during this part of the oscillation cycle, is removing heat in the same direction as the major portion of the heat flow in the system, i.e. that resulting from film desorption and "pumping" toward A. When the film is cooled through its transition temperature, flow toward B begins again and the oscillation cycle repeats. Transition temperature of the film is essentially the same each time the flow begins, since the thickness is the same after each desorption which follows each "cut-off" of the film flow. The gradual rise in salt temperature causes a slight steady film redistribution, causing a slight increase in base line temperature as the oscillations progress.

The metallic thermal link between A and B is a minor factor in accounting for the heat transferred during an oscillation cycle, unless the heat being continuously transferred by it from B to A is of the same magnitude as the heat flow occurring due to the helium adsorption and desorption. Nevertheless, whatever the thermal link, any substantial increase in its conductivity will increase the total thermal linkage between A and B, and one would expect a decrease in the final temperature reached just as desorption from the can ends and oscillations begin.

In the remainder of this chapter, the experimental findings will be presented and discussed in relation to the proposed oscillation mechanism. In addition, the effect on the oscillation phenomena of the variation of background experimental conditions, such as helium gas input and magnetic field, will be discussed. Unless it is mentioned specifically, all the data presented are those obtained with geometrical arrangement 1, but comparisons with the general results obtained with arrangements 2 and 3 will be given where available.

The potential drop across the thermometer T_1 was obtained from the chart record, while the drop across T_2 was read on the potentiometer. The thermometer

resistance values corresponding to these voltages were then converted to absolute temperature readings by comparison with the thermometer calibration. Time was measured directly from the chart, which had a speed of 1" per minute. The reproduction of Fig. 7 shows a typical example of temperature oscillations as seen on the chart record. Fig. 7 may be viewed as a plot of time vs. temperature, where temperature increases toward the left. The shift in oscillations which is seen near the bottom of Fig. 7 is the result of shifting the zero on the chart by changing the bucking voltage (BV) from 0.0605 volts to 0.063 volts. Amplitude of the oscillations shown is 0.0055°K , and the base line temperature near the left of Fig. 7 is 0.642°K . Notice that the oscillations are of the relaxation type, having a slowly increasing base line temperature and constant amplitude until just before the abrupt temperature rise of the can.

Fig. 8 is intended to illustrate the considerable variety of thermal behavior of can and salt that may be obtained by changing conditions of thermal contact for different demagnetization runs. A and B are always in thermal contact through the poorly conducting inconel tube, C. After demagnetization, heat is drained from B to A through C and in addition heat is removed from B

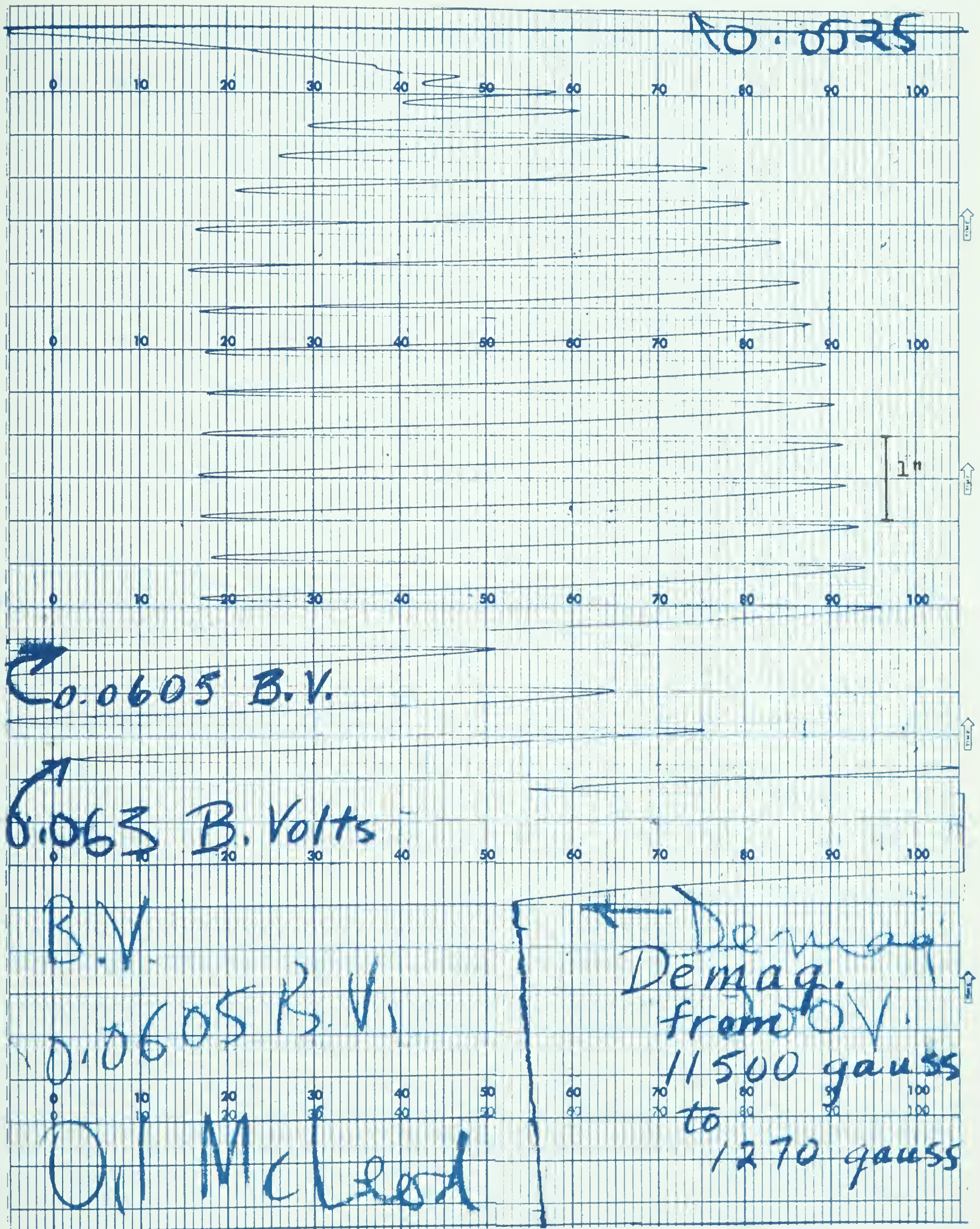


Fig. 7

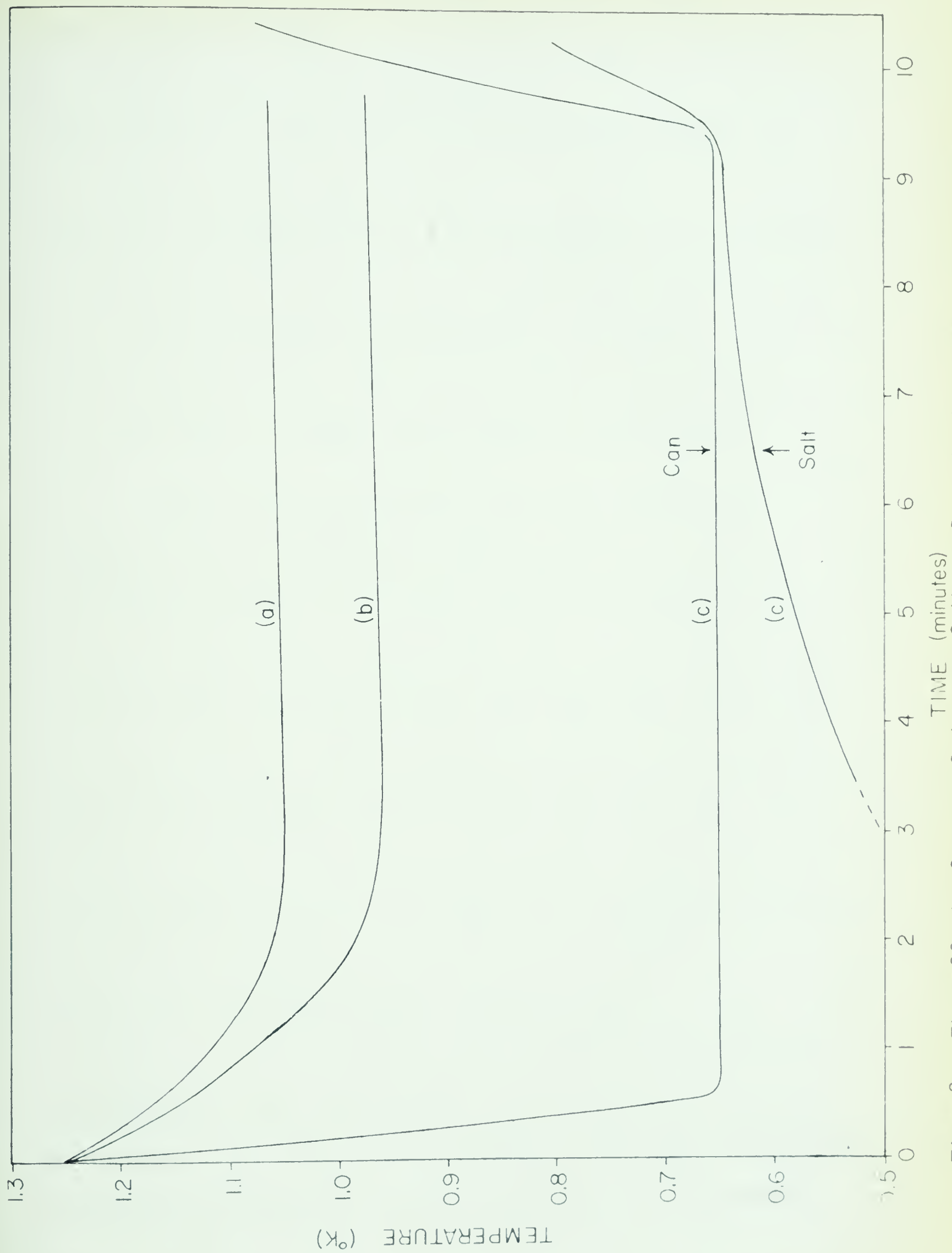


Fig. 8 - The effect of several types of thermal contact on can temperature

because of one of the following thermal links -

- (a) thermal switch, L, in superconducting state, no helium gas or film present
- (b) thermal switch in normally conducting state, no helium gas or film present
- (c) an unsaturated helium film is present on the inside surfaces of A, B, and C. Temperature oscillations are of too small an amplitude to be shown on Fig. 8. Variations due to the state of the thermal switch are also too small to be shown on this graph.

For cases (a) and (b), the salt temperature remains between 0.5°K and 0.6°K for periods as long as three quarters of an hour, but the warming rate is considerably faster for the case (c). When temperature oscillations are taking place, the salt must absorb all the natural heat leak to the whole adsorption chamber, including that which usually warms only the can. Heat leak to the system is larger than usual because of the helium present in all the capillary tubing joined to the top of B. Oscillations are maintained as long as a temperature gradient exists between A and B. When cooling of B can no longer occur, film flowing onto it accumulates, thus raising the transition temperature of the film, and a substantial amount of the heat which was originally removed from B by desorption is redeposited

by adsorption. This is the reason why B warms so rapidly immediately after oscillations cease until a temperature range is reached where the film is more evenly distributed, and warming is caused by natural heat leakage.

A rough calculation estimating the heat removed from B by gas conduction in the tube C shows that this additional amount of heat would not be sufficient to account for the substantial decrease of temperature of B in the case (c) where helium is present in the system. At a temperature of about 0.7°K , the thermal conductivity of inconel is 0.2×10^{-3} watts/ cm-deg (Estermann and Zimmerman, 1952), thermal conductivity of superconducting lead is 1×10^{-2} watts/ cm-deg (Heer, Barnes and Daunt, 1954), and the thermal conductivity of helium gas is 2×10^{-5} watts/ cm-deg (Ubbink and de Haas, 1943). Using the dimensions given in Chapter II, the calculation results are as follows.

<u>Heat Transported from B to A</u>	
Inconel tube	5×10^{-7} watts/ deg.
Lead wire	4×10^{-7}
Helium gas	4×10^{-7}

Evidently the additional thermal contact by the helium gas increases the heat flow between B and A by about 50%, over the case (a). However an increase of heat flow by a factor of perhaps 50, i.e. the difference between

(a) and (b), does not lower the can temperature to the point it reaches in case (c). Therefore, one is led to support the idea that cooling of the can to a temperature as low as 0.6°K is due to desorption of the helium film from its surface.

If helium gas is "pumped" from B to A through C because of the temperature difference caused by demagnetization of the salt, then some gas will be pumped through E into B. However, the pumping speed, or conductance of the fine capillary is much less than the tube C. In this case Poiseuille's law of viscous flow is applicable since the mean free path of the molecules (at 4°K , mean free path of helium is about 0.01 mm. at a pressure of 0.1 mm. Hg) is small compared with the dimensions of the flow channel. Conductance of a long cylindrical tube (Scott, 1959) is given by the formula

$$C = 3.3 \times 10^{-5} \frac{D^4 P}{nL}, \text{ where } C \text{ is conductance}$$

in l./sec., P is pressure in microns, D and L are diameter and length in cm., and n is viscosity in poise. The relative conductance of the tubes C and E is given by a ratio of the quantities D^4/L , which has a numerical value of $9 \times 10^{+5}$. Obviously the gas pumped into the adsorption chamber through E is negligible.

The tubes placed in position C in each of the experimental arrangements 1, 2, and 3 were of the same length but different diameter. Although there is a difference in thermal conductivity of these tubes, the main change in the adsorption chamber is the difference in speed at which the gas desorbed from B can be pumped through C to be deposited on A. The ratios of conductance are as follows -

$$\frac{D_2^4}{D_1^4} = 0.0064 \qquad \frac{D_3^4}{D_1^4} = 15.2$$

Because these conductances differ so widely, one would expect a change not only in rate of pumping but in amount of gas pumped from B to A. Relative to observations on experimental arrangement 1, a substantial increase should be observed in temperature at which oscillations occur in arrangement 2; and in arrangement 3, there should be a substantial decrease in oscillation temperature. Exploratory observations using arrangements 2 and 3 indicate that these predictions were realized. For a constant amount of helium input, the period of the relaxation cycles should also be affected by the pumping speed of the joining tube, but no conclusive evidence on this point has yet been obtained.

The amount of helium admitted to the adsorption chamber is expressed as a number of moles. The usual quantity was something of the order of 300 cc. of gas at a pressure of 10 mm. Hg, at room temperature. Adsorption data is most commonly presented in terms of relative saturation, p/p_0 , but due to the nature of this system, it cannot be accurately estimated. Volume of the system at the helium pot temperature or below was 6.70 cc. with a surface area of 31.5 cm^2 . Volume of the capillary tubing situated at the 4.2°K temperature region in the liquid helium in the dewar was about 1 cc. This tube was in turn joined to the room temperature section of the filling system by a volume of several cc. When a known charge of helium was admitted to this section through stopcock J (of Fig. 5), it was not possible to estimate the amount actually reaching the adsorption chamber proper because the filling line was a significant part of the whole. In addition, there were temperature gradients on the adsorption surface itself.

If the adsorption surface is more or less uniform in temperature, the quantity of helium in the adsorption chamber may be uniquely specified by determining at what temperature the film undergoes a transition. For a particular helium input, there will be a unique thickness of film in equilibrium with the helium vapor, for

every temperature.

A transition temperature for each charge of helium gas was observed, as described in Chapter III, at temperature above 1°K , yet the oscillations were observed below 0.7°K . Oscillations cannot occur unless the helium film flow is choked off because the film transition temperature is exceeded. The only reasonable explanation for this seemingly contradictory measurement of two transition temperatures for the same film is that a drastic redistribution of the helium in the adsorption chamber occurs so that the film on the can after demagnetization is much thinner than before, and therefore its transition temperature is decreased. This is consistent with the idea, mentioned above, that the salt, serving as a cold reservoir, must act as a condensation pump so that most of the film originally on the can is removed by desorption.

Fig. 9 shows the way in which the transition temperature on the can varied with initial helium gas input. The temperature of the adsorption surface was not strictly uniform, since the transitions were observed during warming and cooling of the can which necessarily implied some temperature gradient between A and B (usually about 0.1°K).

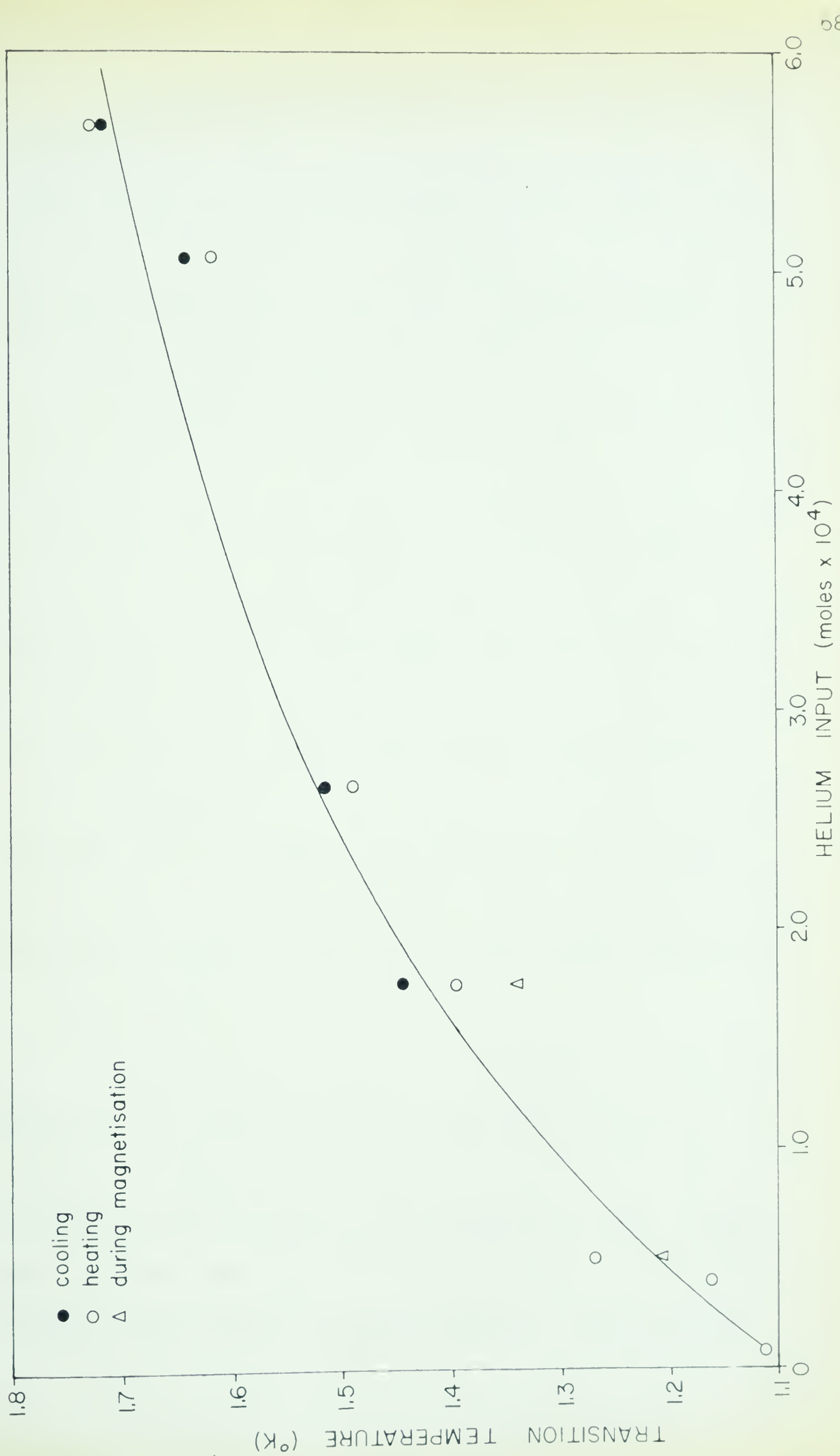


Fig. 9 - Onset of superfluidity as a function of helium input

The observations for Fig. 9 were made in three different ways, each of which caused the appearance of a film transition at a slightly different temperature. As mentioned in Chapter III, transitions were observed when the system was warming after a demagnetisation. The warming curve for the can, shown at the left of Fig. 10 has been traced directly from the chart record. Warming rate of the can was increased when the heater was turned on. Below the transition temperature of the film, the heater current also warmed the salt as a result of the thermal contact afforded by the film joining B and A. Above the transition temperature, the film was immobile and therefore only the can was warmed, at a much faster rate than before. Temperature corresponding to this change of slope could be determined to about three-figure accuracy. When the heater was switched off, the can would cool to the transition temperature and more slowly thereafter because of the heat of adsorption deposited by the film flowing onto B from A.

During magnetization of the salt, the heat of magnetization would sometimes cause the can to warm through the superflow onset temperature for the film thickness present there. The temperature scale is the same for both right and left sides of Fig. 10, but the diagram at the right is placed higher on the ordinate

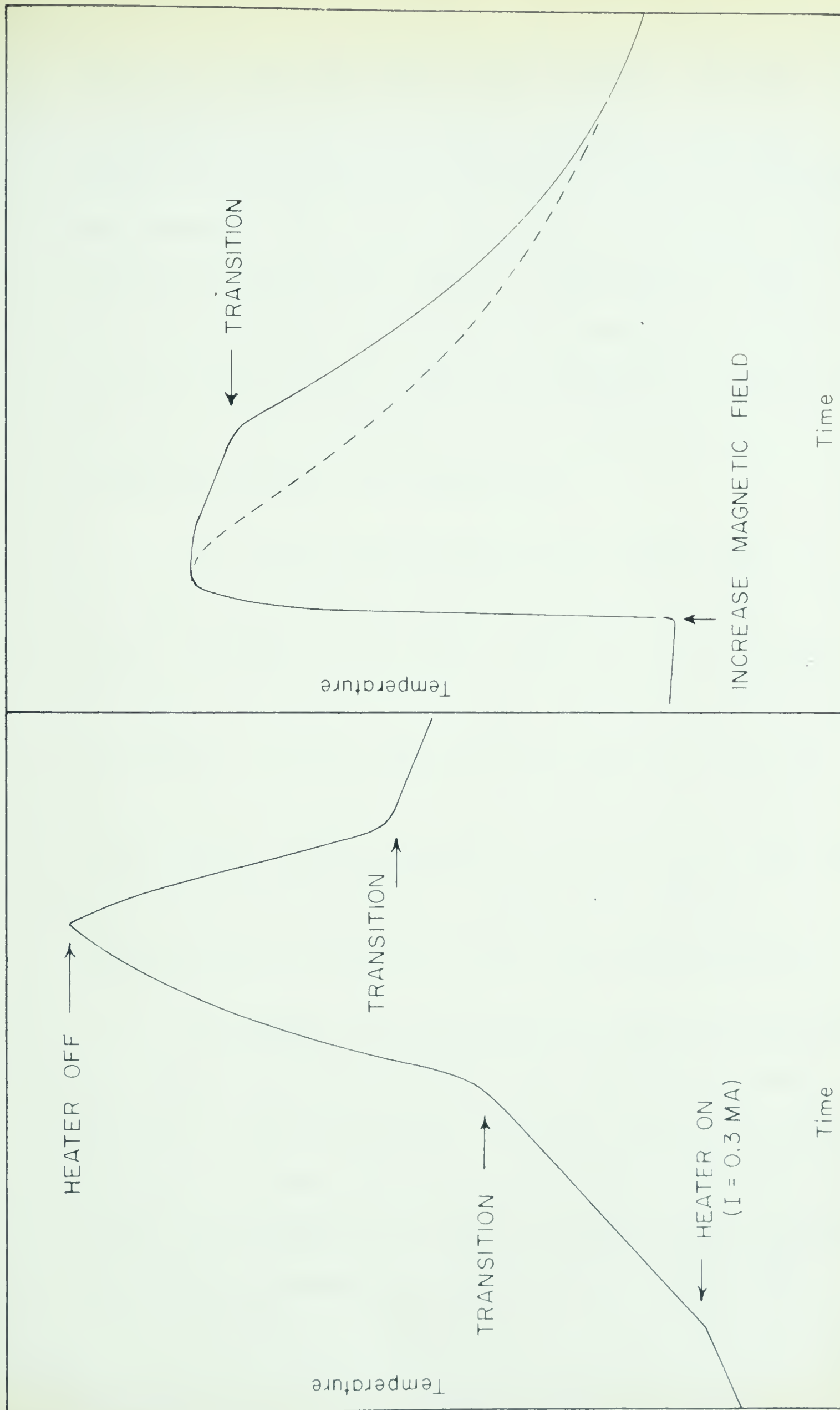


Fig. 10

compared to the left side of the diagram. As shown by the solid line at the right of Fig. 10, the can cooled at different rates above and below the transition temperature. The cooling rate is increased below the transition temperature, which is the reverse of the situation discussed in the previous paragraph. This is explained by noting that the temperature gradient is reversed in this situation, and so film flows from B to A below the transition temperature.

The final distribution of the helium film on the adsorption surfaces should alter as the helium input varies, so that as the film on the can becomes thinner, the base line temperature of the oscillations decreases. The effect of helium input on can temperature during oscillations is shown in Fig. 11. Since the effect is rather small, the most probable explanation is that the major factor influencing can temperature during oscillations was geometry of the adsorption chamber.

The first one or two cycles of the oscillations were often observed to have an amplitude about twice as large as the average for the later cycles. The base line of the first one or two cycles was also placed at a much lower temperature than the cycles which followed. Two possible explanations come to mind. Either the

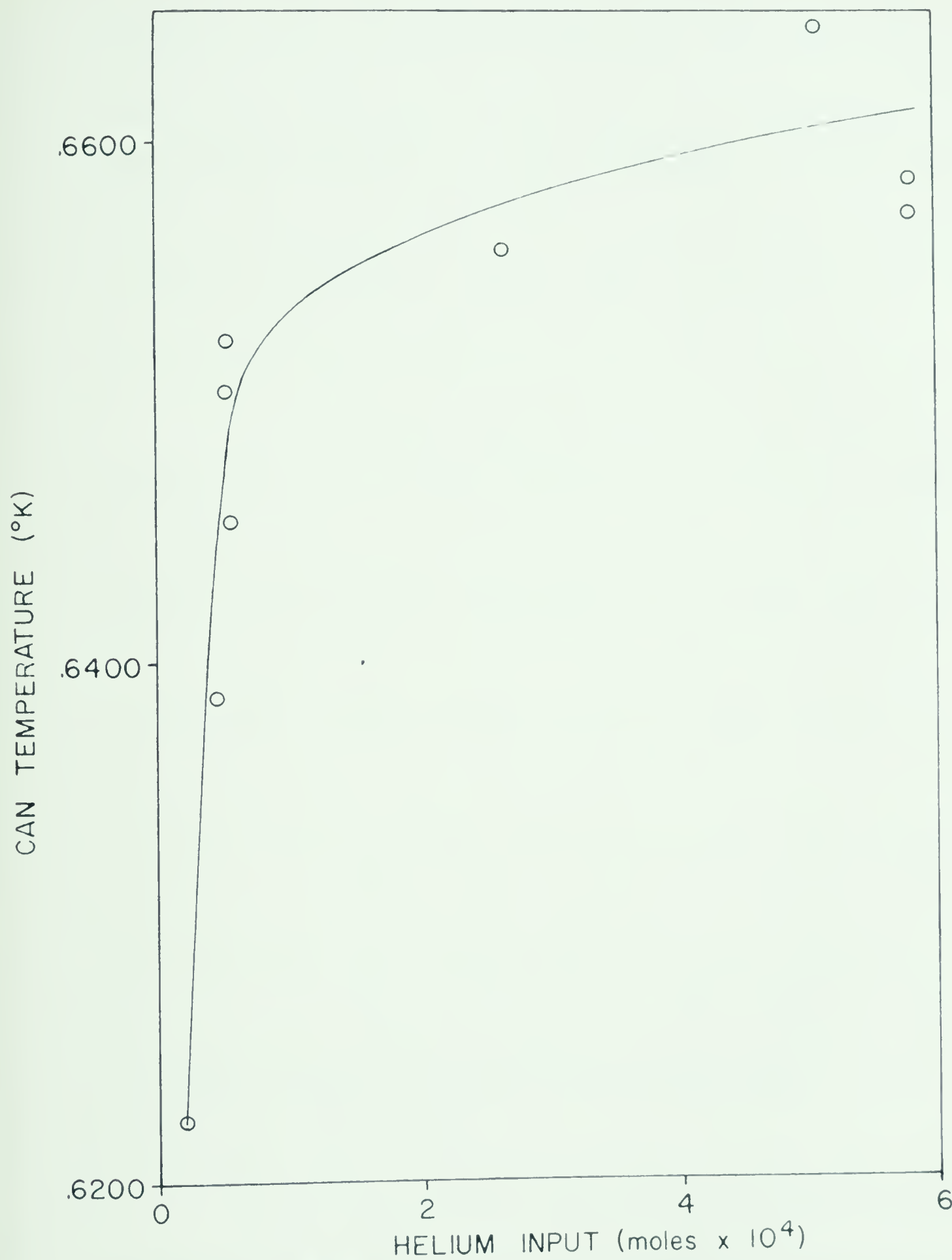


Fig. 11 - Dependence of the base line temperature of the oscillations on film distribution

amount of film moved in the first cycles was twice as great as that moved in the later cycles, or if the amount of film moved was nearly the same for every cycle, almost all film was stripped from the can immediately after demagnetization. Since the heat of adsorption during formation of the first and second statistical layers is relatively large, these layers may have been the ones formed during the first cycles.

It is easily seen from Fig. 7 that the amplitude of the oscillations was essentially constant, except, of course, when the oscillations were nearly ended. Not all oscillations observed were as even and well defined as the set shown presumably because of some unspecified external interference. Nevertheless, it was possible to make a satisfactory estimate of average amplitude for every set of oscillations, even though there were some individual cycles of irregular shape and amplitude. The information obtained, presented in Fig. 12, shows some evidence that amplitude of the oscillations was increased by introduction of more helium into the system.

There are several observations which indicate that the period of oscillation increases with helium input, but the available data is not conclusive enough

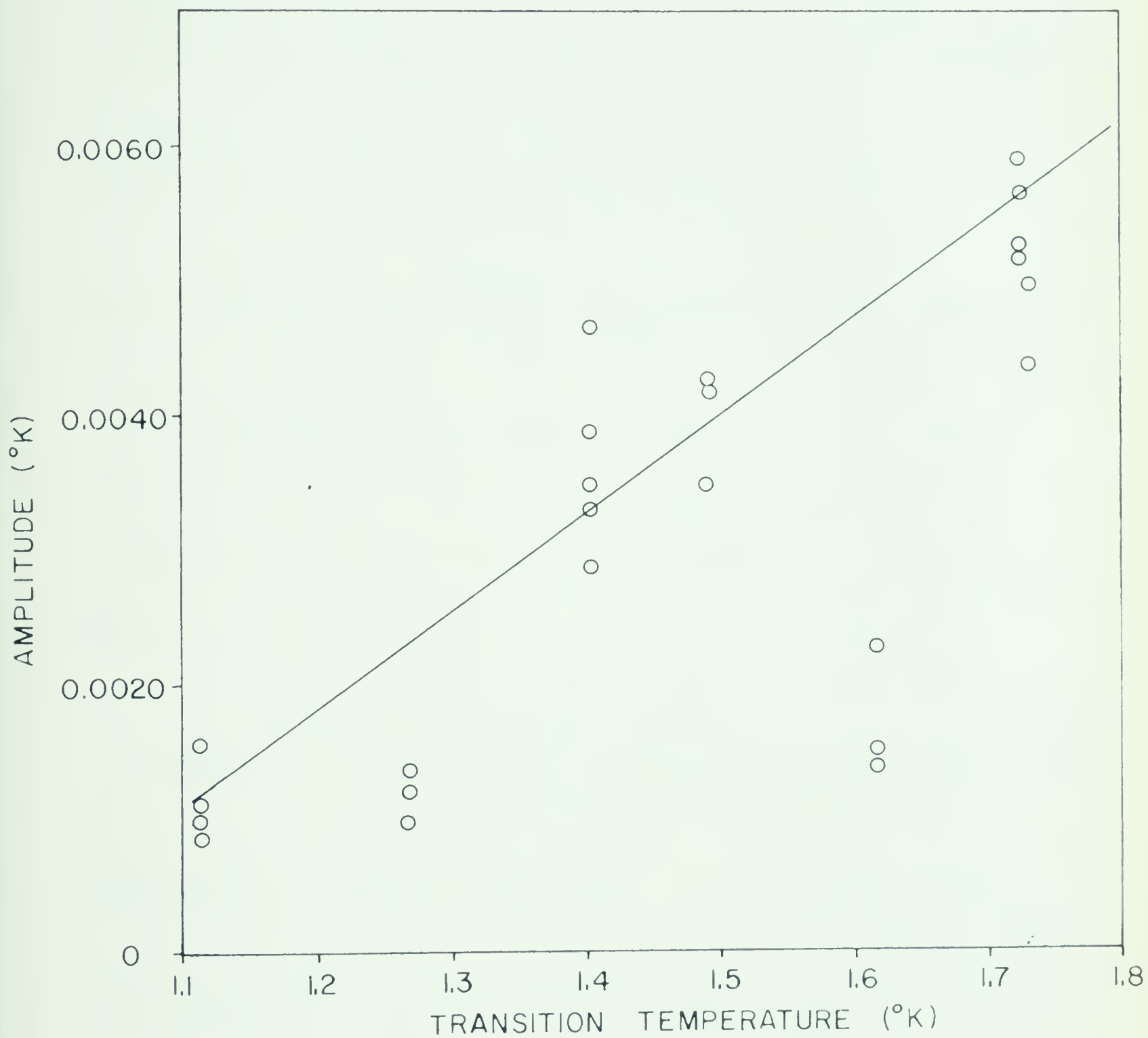


Fig. 12 - Dependence of average oscillation amplitude on the unsaturated film thickness

to present at this time. Periods as small as 10 sec. and as large as 26 sec. have been produced during the course of these experiments. Time elapsed between demagnetization and the abrupt temperature rise (because of insufficient temperature gradient between can and salt) has been found to depend on the original amount of helium gas admitted to the system. Fig. 13 illustrates that especially for a small amount of helium input, time elapsed decreases as input increases. Data for arrangement 1 is denoted by open circles, that for arrangement 2 by closed circles. For the larger amounts of helium input, there may be increased heat leak to the system due to the presence of a thicker helium film on the inner surfaces of the fine capillary, E.

Even though the salt temperature was slowly rising toward the can temperature, oscillations were maintained at a constant period and amplitude for most of the duration of each set observed. To study the possible changes of oscillation behavior which would result from a change in the salt temperature after demagnetization, the salt was magnetized to fields less than the maximum available field of 11,500 gauss. It was found that the base line temperature shifted to a slightly lower value when the salt was demagnetized from a relatively small

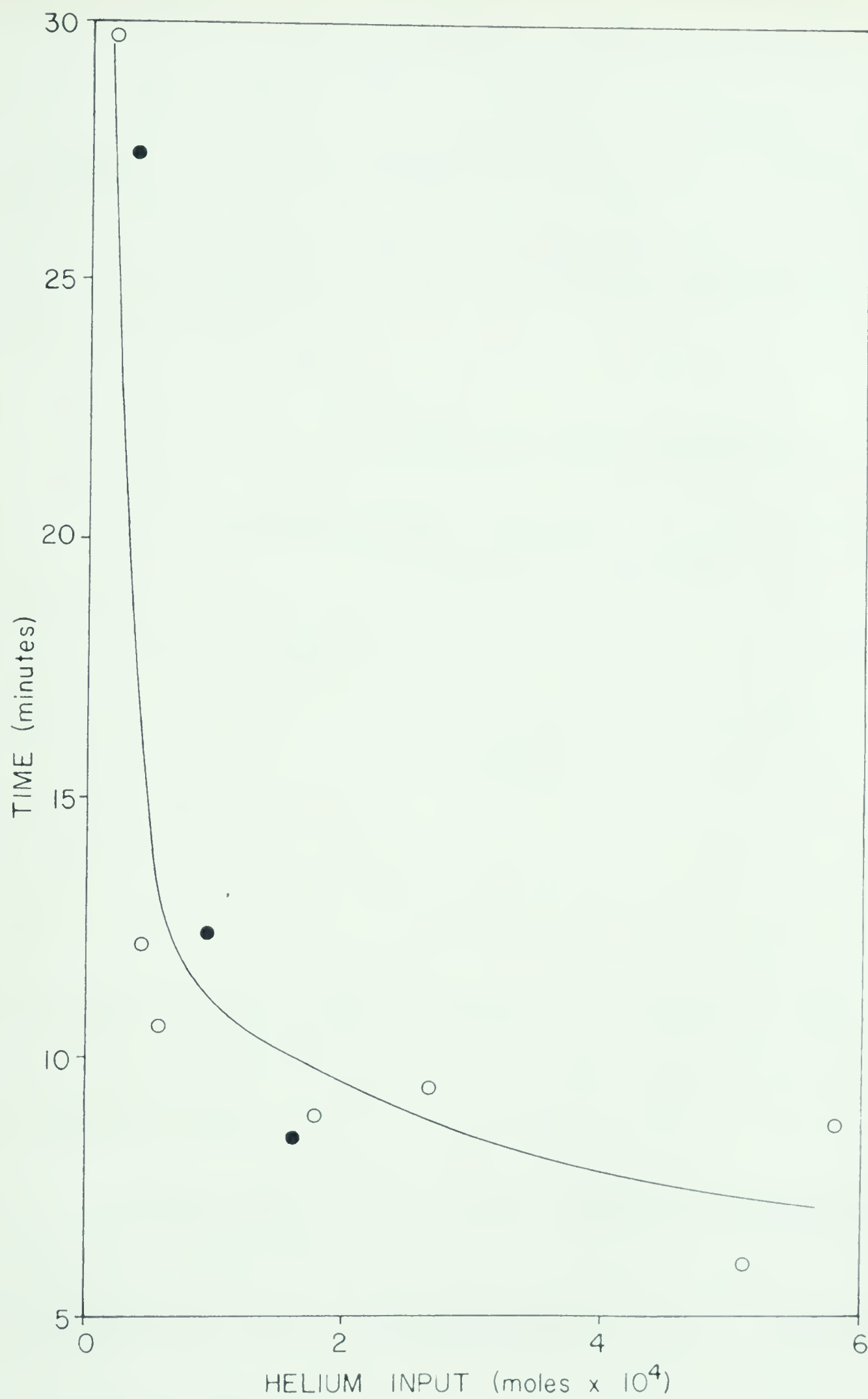


Fig. 13

field, so apparently the major factor influencing base line temperature was geometry.

The results listed in Table I indicate a decrease in amplitude (with the exception of III of Group 1) caused by demagnetization from a smaller field. The demagnetizations of each group were performed for a constant helium input.

TABLE I

	Field before Demagnetization		
	11,500 gauss	7,700 g.	6,500 g.
	OSCILLATION AMPLITUDE ($^{\circ}$ K)		
<u>Group 1</u>			
I	0.0028		
II	0.0028		
III			0.0041
IV		0.0016	
V		0.0017	
<u>Group 2</u>			
I	0.0012		
II		0.0006	
III			0.0003

These results are rather difficult to interpret and there may be some unknown experimental conditions which influenced them.

Regardless of the initial magnetic field, the temperature measured by the salt thermometer after demagnetization was always approximately the same. According

to a curve of final temperature vs. initial magnetic field obtained by de Klerk (reproduced by Zemansky, 1957), for demagnetization of potassium chrome alum from a temperature of 1.17°K , the final temperature of the paramagnetic salt was as follows.

<u>Initial Magnetic Field</u>	<u>Final Temperature</u>
11,500 gauss	0.05°K
7,500	0.15

Probably these temperatures were actually attained in the interior of the salt capsule, but thermal contact between the tube A and the thermometer may have been poor below 0.5°K because of the baking varnish insulation. Apparently the low temperatures which the salt should have reached could not be observed except by the introduction of a more elaborate way of attaining thermal contact.

It was observed that the time occupied by a set of oscillations decreased by a factor of nearly four when the magnetic field around the salt before demagnetization was reduced by a factor of two. This is consistent with the conclusions above. If the salt does not reach so low a temperature after demagnetization, then it requires less time for it to warm to the can temperature, which causes the oscillations to end sooner than before.

Changing the state of the thermal switch from superconducting to normal increased the heat transported by the metallic link between B and A by a factor of about fifty. Observations of the effect on the oscillations of changing the thermal switch should indicate the relative importance to the oscillation mechanism of the metallic thermal link in comparison to the thermal contact provided by the helium vapor and film. As discussed in Chapter III, the thermal switch was "on" at a field of 1270 gauss, and "off" at a field of 660 gauss. Either the switch was changed during a set of oscillations, or two sets of oscillations were observed, under identical conditions, with the exception that the thermal switch condition was different for each set.

Results available at the present time are summarized in Table II, and the order in which switching was done may be found by reading from left to right. The number in parentheses indicates the number of cycles over which the amplitude was averaged. Helium input was constant for each group of observations. A study of Table II shows that (with the exception of VI of Group 2) there is a tendency to increased oscillation amplitude when better thermal contact by metallic linkage is produced between can and salt.

TABLE II
Effect of Thermal Switching on Average Amplitude

<u>Group 1</u>	<u>Switch On</u>	<u>Switch Off</u>	<u>Switch On</u>
I	0.0028°K (16)		
II		0.0027°K (15)	
III	0.0037 (6)	0.0031 (13)	
<u>Group 2</u>			
I	0.0044 (15)		
II		0.0022 (13)	0.0045°K (3)
III		0.0037 (19)	
IV		0.0024 (10)	0.0035 (6)
V		0.0040 (8)	0.0042 (7)
VI	0.0026 (11)	0.0033 (11)	
VII	0.0046 (9)	0.0038 (9)	

An amplitude increase in the oscillations may mean that film flow was not choked off so quickly because heat was being drawn away from the can and film by thermal conduction through the lead wire, but in this case one would have to suppose that heat removed from the can by extra thermal conduction was less than the heat of adsorption deposited on the can by the extra film flowing onto it. Heat should be removed quickly during the part of the cycle when the film was choked off but no change in period has been observed. The results of Table II are not conclusive evidence that oscillation amplitude changes with the thermal switch action, since the average amplitude observed is not accurately reproducible from one set of oscillations to the next. However,

it can definitely be stated that the effect of the thermal switch was not large, thus lending support to the conclusion that the major factor in the heating and cooling of the can is adsorption and desorption of helium from its surface. It is hoped to undertake a more detailed study of the effect of thermal switching by experiments with arrangement 3, which has a switch containing more lead, of higher purity.

There is one aspect of the temperature oscillation phenomena which is not understood. The pressure in the helium filling system, as measured at room temperature, increased during a set of oscillations in the manner illustrated in Fig. 14. The letter A denotes the time when exchange gas was pumped from the experimental chamber, B denotes demagnetization of the salt, and C marks the time when oscillations ended. On some occasions (see top curve) the pressure began to increase slowly as soon as the exchange gas was pumped from V_2 , while at other times (see bottom curve) the pressure rise began only after the cooling of the system. The pressure fell toward its steady value during magnetization when the oscillations stopped, although the pressure rise was occasionally noticed to continue for a few minutes after oscillations ceased.

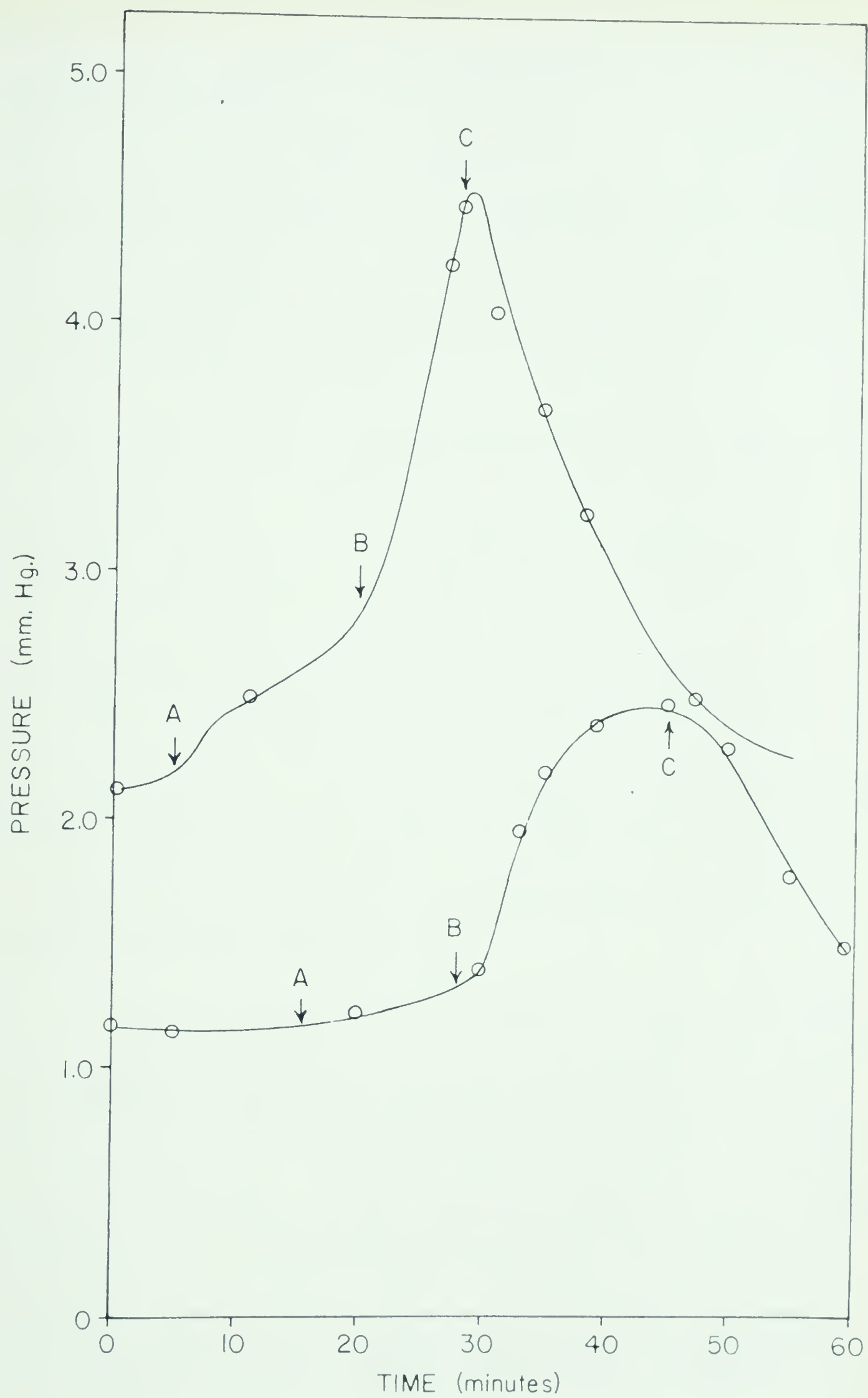


Fig. 14

It would be reasonable to assume that a temperature increase at the low temperature end of the filling system would cause a pressure increase in the room temperature part of the system because of a gas redistribution and a probable desorption of gas from some of the surfaces in the system. A pressure increase of the order of 15% has been observed at room temperature when the adsorption chamber was warmed from 1.2°K to 4.2°K . Warming due to magnetization did not produce any significant pressure increase. In view of this "normal" behavior, it is difficult to understand the observations shown in Fig. 14. A small part of the pressure rise may be due to removal of exchange gas from that part of the pumping line P_2 which passed through the helium pot. Heat conducted along the tubes in the line P_2 from the bath at 4.2°K would then cause a temperature rise in that part of the capillary tube and in the fine capillary, E.

This phenomenon may be related to conditions encountered during the course of experiments on vapor pressure of He^3 - He^4 mixtures (Sommers, 1952). A sample chamber, immersed in a He^4 bath was connected to a large reservoir at room temperature through a small diameter stainless steel filling tube. At low temperatures, using pure He^4 in the system, there was a pressure

difference between the bath and the helium sample in the holder, such that at 1.180°K , excess pressure inside the sample holder was 0.3 mm. Hg. The pressure difference dropped as the logarithm of the bath pressure and was negligible at 1.55°K . Sommers' investigations show that the pressure differences were not due to the thermomolecular pressure drop, or depth of immersion of the sample holder in the bath, or gas evaporating from a superfluid film, or a temperature difference between bath and sample holder. The true reason for the pressure difference was not understood, but it decreased when a larger filling tube was used.

If the pressures shown in Fig. 14 corresponded directly to the conditions in the adsorption chamber, some of them would indicate a pressure excess over saturated vapor pressure for the temperatures at which the oscillations occur. As in Sommers' experiments, this pressure excess increases steeply as the temperature of the sample chamber decreases. Further experiments are necessary to investigate these effects.

CHAPTER V

CONCLUSIONS

The existence of temperature oscillations under the conditions discussed in this thesis is interesting in itself as a phenomenon due to the combined effect of helium gas adsorption and superfluid flow in He II. The fact that oscillations have been produced by approaching the problem in the manner suggested in the introduction indicates that this work has helped to account for the unusual thermal behavior observed in some adiabatic demagnetization experiments, thereby accomplishing one of the original aims of this project. From a broader viewpoint, this experiment is useful because the implications of these results provide additional information about the nature of the unsaturated helium film.

As expected, a decrease in film thickness resulted in a decrease in transition temperature, the lowest temperature for onset of superflow observed in this experiment being about 1.1°K (see Fig. 9). Prior to this work, the lowest transition temperature observed was about 1.3°K (see Fig. 2). However, during the oscillations, a film transition has occurred on a number of occasions at temperatures as low as 0.65°K (even lower

transitions were indicated by preliminary findings with experimental arrangement 3), which suggests that the films produced were much thinner than any films observed previously in this connection. All the transition temperatures mentioned above were found by observing the onset of superflow. The last method of observing a transition is unique in that the transition occurs repeatedly (once per cycle), without an external method of triggering the transition (such as heating part of the adsorption surface). Long and Meyer (1955) estimated that the onset of superfluidity which they observed at 1.3°K corresponded to a film less than two statistical layers in thickness. Recent evidence indicates that, contrary to Long and Meyer's assumptions, the first layer is not four times as dense as other layers (as discussed in Chapter I), so that their estimate of two layers for the thickness corresponding to a transition at 1.3°K must be in error. Assuming that the first layer is immobile - it is probably solid and formed at a very low relative saturation, as discussed in Chapter I - then at least part of the second layer must be formed to allow film movement at 0.65°K , so a film that is mobile up to 1.3°K must contain a greater average film thickness above the first layer. The present experiment does not concur with the theoretical predictions of

Hecht (1958) that no film transition is possible in a film less than $1\frac{1}{2}$ layers thick, with a corresponding onset temperature of about 1°K . It may be that the weakness in this theory lies in the arbitrary choice of 1°K as the point below which one of the parameters used in the theory takes on a ~~non-zero~~ value, whereas it is chosen to be zero above 1°K .

Further evidence that the film on the can during the temperature oscillations is extremely thin is available in the observations of the relatively large amplitude of the first cycles of a set of oscillations. It is known that the heat of adsorption for a film several layers thick is only a little greater than the heat of vaporization, but the heat of adsorption for the first layers is considerably greater than this. A large amplitude oscillation may indicate the heat of adsorption deposited on the surface of the can during the formation of the first one or two film layers.

The adsorption-desorption cycle which causes the temperature oscillations in this experiment may be regarded as an exaggerated form of the heat transport cycle suggested by Long and Meyer (1952b, 1955). In the case of saturated film flow, it is known that film which evaporates is replenished by superfluid flow from the

He II bath, so that there is a continuous "cycle" of film evaporation, recondensation in the bath, and film replacement by superfluid flow, especially if there is a slight temperature gradient between the bath and the surface on which the saturated film is present. Evidently the same heat transport cycle is found in unsaturated film phenomena, with recondensation taking place at the region of lower temperature on the adsorption surface.

It is of considerable interest in understanding the nature of the lambda transition to know if the transition is discontinuous. It has generally been assumed that the bulk liquid lambda transition is sharp, but, as mentioned in Chapter I, no work has been undertaken to determine if a similar sharpness exists in the unsaturated helium film transition. In the first experiments directed toward observation of transition sharpness, Keesom and Miss Keesom (1935) concluded that the specific heat jump in the bulk liquid occurs abruptly or within a temperature interval smaller than 0.0002°K . More recently, it has been possible to measure the specific heat of liquid helium near the lambda point with a resolution of several millionths of a degree (Fairbank, Buckingham, and Kellers, 1958). These results indicate a sharp transition having a logarithmic singularity in the specific heat at the lambda point.

From a thermodynamic viewpoint, the lambda transition bears some resemblance to a second order transition, which must satisfy the conditions set forth by Ehrenfest, i.e. at the transition there is a discontinuity in the second derivatives of the Gibbs function, namely C_p , $(dv/dT)_p$, $(dv/dp)_T$ (the usual thermodynamic notation is employed). For a second order transition, the discontinuity in the specific heat is finite, and there is no latent heat. However, it is not experimentally certain whether there is a latent heat for the He I - He II transition or if it is just very small, and the very high peak in the specific heat curve is usually considered to indicate an infinite discontinuity. Even if the lambda transition is ideally discontinuous, it is still possible that statistical fluctuations may limit the actual sharpness of the transition regardless of the possible experimental accuracy that could be obtained. The nature of the lambda transition has been discussed in detail by Keesom (1942) and more recently by Pippard (1957).

The specific heat curves presently available for unsaturated helium films (Frederikse, 1949; Champeney, 1958) do not have the typical sharp peak and discontinuity that appears in the bulk liquid specific heat curve. Without further evidence, subsequent comment

(Long and Meyer, 1953; Daunt and Smith, 1954) has assumed that the specific heat anomalies in these curves are not sharp but actually the experimental data is not sufficiently accurate to allow any final conclusions on this point.

Although the specific heat maxima are not sharp, onset temperatures for superflow measured by Long and Meyer (1952) were sharp to about 0.001°K . All of the work described in this thesis depends on a certain sharpness in the transition. In Fig. 15, part of a typical set of oscillations is shown, and it can be seen that deviations of the experimental curve from a smooth base line are not more than 0.0002°K . These results suggest that a transition has been observed which was about one order of magnitude sharper than any transition observed in the unsaturated helium film up to the present time.

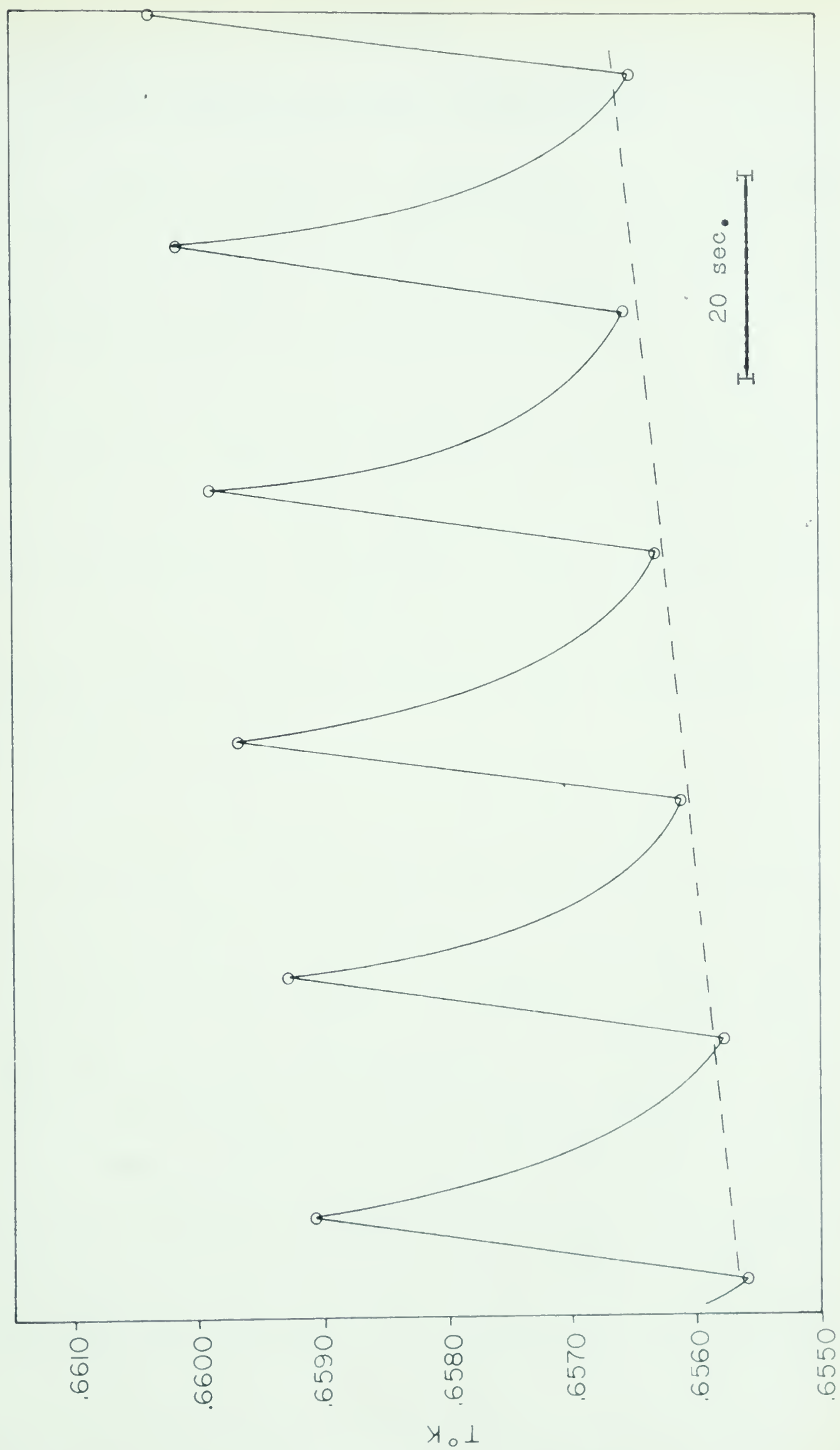


Fig. 15 - An illustration of the sharpness of the film transition

APPENDIX I

THE CARBON RESISTANCE THERMOMETERS

Carbon resistance thermometers are very suitable for use at liquid helium temperatures because of a high sensitivity to small temperature changes, i.e. $\frac{1}{R} \left(\frac{dR}{dT} \right)$ increases rapidly as temperature decreases. All thermometers used in the temperature oscillation experiment were Allen-Bradley (Ohmite), $\frac{1}{2}$ watt, nominal 10 ohm carbon resistors. This type of resistor has already given satisfactory performance in several other experiments (Clement et al, 1953; Pearce, Dillinger and Markham, 1956; Nicol and Soller, 1957; Markham, Netzel, and Dillinger, 1957).

It was found in all these investigations that the resistance - temperature variation of the carbon resistors could be represented by an equation of the form $(\log R)/T = A + B (\log R) + C (\log R)^2$. This semi-empirical form was first suggested by Clement and Quinell (1952). Clement et al (1953) reported excellent agreement between temperatures calculated from the above equation and those obtained in their calibrations, using magnetic susceptibility measurements from 0.15°K to 1.3°K , and comparison with helium vapor pressure in the range from

1.25°K to 4.2°K. After warming to room temperature, reproducibility of temperatures calculated from resistance values was found to be $\pm 0.2\%$ above 1°K and $\pm 1\%$ below 1°K. Nicol and Soller (1957) calibrated carbon resistors between 4.2°K and 1°K and found that observed resistance values below 1°K compared with the predictions of the calibration equation.

For the work described by this thesis the thermometers were calibrated against the T_{55E} temperature scale, by making a series of measurements of resistance values of the thermometers in exchange gas contact with the helium pot. The manostat in the helium pot pumping line was used to control the pressure, and thus to hasten the approach of thermal equilibrium necessary for obtaining each point on the calibration curve.

A least squares curve fit to an equation of the form $y = A + B (\ln x) + C (\ln x)^2$, where $y = (\ln R)/T$ and $x = R$, was calculated using the LGP-30 computer at the University of Alberta Computing Center for each thermometer calibration. The computer was also used for a 1200-value tabulation of temperature corresponding to resistance according to the coefficients A, B, and C obtained with the curve fit calculation for the thermometer T_1 used with experimental arrangement 1, and also for T_1

used with arrangements 2 and 3. The coefficients used in these tabulations are given in Table III.

TABLE III

	<u>A</u>	<u>B</u>	<u>C</u>
Arr. 1	2.622596	-1.524962	0.2587947
Arr. 2 & 3	10.76426	-4.657323	0.5563838

There is a marked difference in the two sets of coefficients. The thermometer used on the second copper canister (for arrangements 2 and 3) was chosen from a different lot of resistors and proved to be much less sensitive than the previous thermometer. For the range of temperature covered by the calibration, the difference between the experimental data and corresponding points on the fitted curve was between 0.5% and 4% for the second thermometer, whereas for the first thermometer agreement between the curve and the experimental points was not worse than about 0.03%.

Temperatures below the minimum helium pot temperature of 1.2°K were calculated from the extrapolation of the calibration curve. Other facilities for measuring temperature below 1°K directly were not available in this laboratory so it is necessary to rely on the results of other investigators and assume that the extrapolation is valid. Considering this limitation,

it may be true that the temperatures corresponding to the measured resistance values do not accurately indicate absolute temperature values, but temperature differences measured with these thermometers are sufficient for the present purposes. Since the curve fit for the second thermometer was rather poor, quantitative data obtained with it is of doubtful value, but certainly the thermometer is good enough to allow comparison of behavior of arrangements 2 and 3 with that of arrangement 1.

Although it is necessary to recalibrate carbon thermometers after each warming to room temperature in calorimetry experiments (Markham, Netzel, and Dillinger, 1957; Manchester, 1959), such calibration was not required for this work. After warming to room temperature it was found that resistance and temperature values varied from the calibration curve by no greater amount than the original calibration data.

Since some of the temperature oscillations took place when the adsorption chamber was in a magnetic field of the order of 1000 gauss, the possibility of resistance change in the thermometers had to be investigated. Within the experimental error imposed by the fact that as the helium pot was pumped there was a very small but steady decrease of temperature while pumping at full

speed, no resistance change in the thermometers was found at any magnetic field. This is in accordance with the resistance change predicted by the Clement and Quinell (1952) equation, concerning the expected linear relationship between R and H^2 , at constant temperature.

$$\frac{d(R/R_o)}{d(H^2)} = (5.5 + 1.8 \log_{10} R_n) T^{-1.5} \times 10^{-5} \quad (1)$$

where R_n = nominal resistance at room temperature

T = absolute temperature

R_o = zero field resistance at T

H = magnetic field in kilogauss

At $T = 0.65^{\circ}\text{K}$ (a typical oscillation temperature), the value of the right hand side of equation (1) is 0.00014. Therefore in a magnetic field of one kilogauss, a resistance increase of 0.014% is expected. Considering the resistance of T_1 (in arrangement 1) at the oscillation temperature, this change might cause an error in the temperature of 0.002%. These changes are not significant to the accuracy of the data in this experiment, so no corrections need be applied to account for resistance increase in the thermometers situated in a magnetic field.

APPENDIX II

THE OIL MCLEOD PRESSURE GAUGE

The oil McLeod gauge, shown in Fig. 16, was first used by Nester (1957). This gauge and an oil manometer were the means of determining pressure in the helium gas filling system. The oil McLeod gauge consists of a small diameter glass tube closed at the top and joined at the bottom to a section of larger diameter glass tubing. The glass section is surmounted by a fairly heavy steel tube, centered in the outer glass casing by teflon washers, which also facilitate movement in and out of the liquid, a low vapor pressure oil (Apiezon B). Pressure readings in the range 0.1 to 30 mm. of mercury are obtained by lowering the plunger with the magnet, until the top of the capillary, A, is just at the level of the oil meniscus. Pressures corresponding to the height difference, Δh , are found in a manner slightly different from the conventional McLeod gauge calibration, since the volume of the capillary tube (cross-sectional area σ) forms a significant proportion of the whole volume of the gauge. Using the notation of Fig. 16

$$P_1 V_1 = (\Delta h + P_1) \sigma \Delta h \qquad P_1 = \frac{\sigma (\Delta h)^2}{V_1 - \sigma (\Delta h)}$$

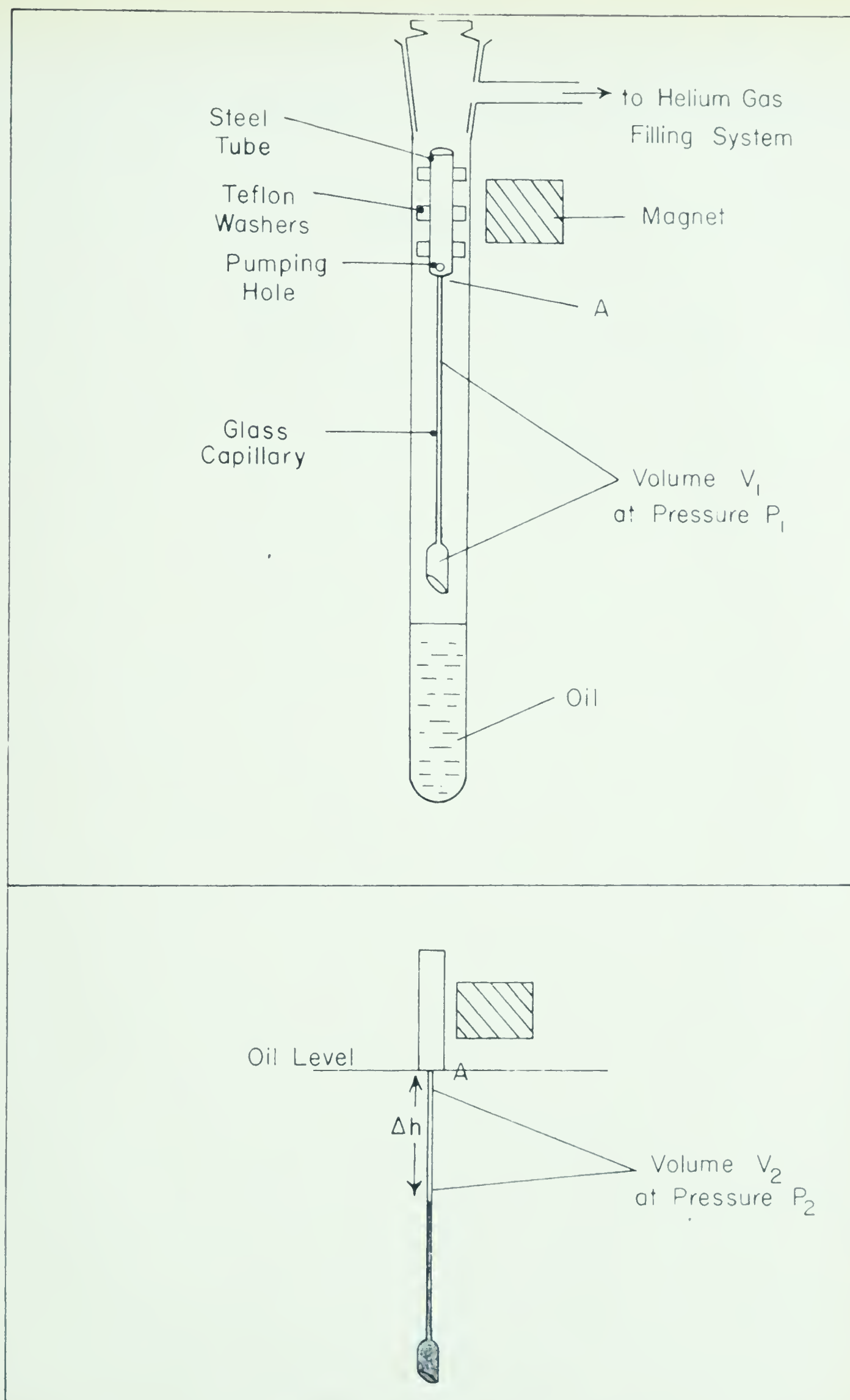


Fig. 16 - The Oil McLeod Pressure Gauge

A brief summary of the calibration for the particular gauge used in this experiment is given in Table IV.

TABLE IV

<u>Δh</u>	<u>P</u>
10 mm.	0.0215 mm. Hg
50	0.621
90	2.393
130	6.095

REFERENCES

- ALLEN, J. F., and JONES, H., 1938, Nature, 141, 243
- ASTON, J. G., MASTRANGELO, S. V. R., and TYKODI, 1955,
J. Chem. Phys., 23, 1633
- ATKINS, K. R., 1954, Can. J. Phys., 32, 347
- ATKINS, K. R., and SEKI, 1958, Proc. V Internat. Conf.
on L. Temp. Phys. & Chem., Madison, Wisconsin, p.27
- BOWERS, R., 1953, Phil. Mag., 44, 485
- BOWERS, R., BREWER, D. F., and MENDELSSOHN, K., 1951
Phil. Mag., 42, 1445
- BREWER, D., and MENDELSSOHN, K., 1953, Phil. Mag., 44, 340
- BRUNAUER, S., EMMET, P. H., and TELLER, E., 1938
J. Amer. Chem. Soc., 60, 309
- CHAMPENEY, 1958, Proc. V Internat. Conf. on L. Temp. Phys. &
Chem., Madison, Wisconsin, p. 29
- CLEMENT, J. R., 1956, U. S. Naval Research Laboratory Report
- CLEMENT, J. R., and QUINNELL, E. H., 1952, R. S. I., 23, 213
- CLEMENT, J. R., QUINNELL, E. H., STEELE, M. C., and HEIN, R. A.,
1953, Rev. Sci. Instr., 24, 545
- CLEMENT, J. R., LOGAN, J. K., and GAFFNEY, J., 1955
Phys. Rev., 100, 743
- DAUNT, J. G., and MENDELSSOHN, K., 1939, Nature, 143, 719
- DAUNT, J. G. and SMITH, R. S., 1954, Rev. Mod. Phys., 26, 172
- DUGDALE, J. S., and MACDONALD, D. K. C., 1957, Can. J.
Phys., 35, 271
- DUGDALE, J. S., MACDONALD, D. K. C., and CROXON, A. A. M.,
1957, Can. J. Phys., 35, 502
- ESTERMANN, I., and ZIMMERMAN, J. E., 1952, J. Appl. Phys.,
23, 578
- FAIRBANK, W. M., BUCKINGHAM, M. J., and KELLERS, C. F.,
1958, Proc. V Internat. Conf. on L. Temp. Phys. &
Chem., Madison, Wisconsin, p. 50

- FRANCHETTI, S., 1956, Nuovo Cim., 4, 1504
- FRANCHETTI, S., 1957, Nuovo Cim., 5, 1266
- FREDERIKSE, H. P. R., 1949, Physica, 15, 860; 1950, Thesis, Leiden
- FRENKEL, J., 1940, J. Phys. Moscow, 2, 365
- FRENKEL, J., 1946, Kinetic Theory of Liquids, Oxford Univ. Press, New York, p. 332
- HAM, A. C. and JACKSON, L.C., 1957, Proc. Roy Soc., A, 240, 243
- HECHT, C. E., 1958, Physica, 24, 1023
- HEER, C. V., BARNES, C. B. and DAUNT, J. G., 1954, Rev. Sci. Instr., 25, 1088
- HOBSON, J. P., 1959, Can. J. Phys., 37, 300
- HULL, R. A., 1947, Rep. Internat. Conf. on Fundamental Particles and Low Temps., - July, 1946, Vol. II, p.72
- JACKSON, L. C. and GRIMES, L. G., 1958, Phil. Mag. Supp., 28, 435
- KEESOM, W. H., 1942, Helium, Elsevier
- KEESOM, W. H., and KEESOM, Miss A. P., 1935, Commun. Leiden No. 235d; Physica, 's-Grav., 2, 557
- KEESOM, W. H., and SCHWEERS, J., 1941a, Physica, 8, 1020
- KEESOM, W. H., and SCHWEERS, J., 1941b, Physica, 8, 1032
- KISTEMAKER, J., 1947, Physica, 13, 81
- LONDON, H., 1939, Proc. Roy. Soc., A 171, 484
- LONG, E., and MEYER, L., 1952a, Phys. Rev., 85, 1030
- LONG, E., and MEYER, L., 1952b, Phys. Rev., 87, 153
- LONG, E., and MEYER, L., 1953, Phil. Mag. Supp., 2, 1
- LONG, E., and MEYER., L, 1955, Phys. Rev., 98, 1616

- MANCHESTER, F. D., 1959, Can. J. Phys., 37, 989
- MANCHESTER, F. D., and BUCKLEY, Adele, 1960, Proc.
VII Internat. Conf. on L. Temp. Phys., Toronto
- MARKHAM, A. H., NETZEL, R. G., and DILLINGER, J. R.,
1957, Rev. Sci. Instr., 28, 382
- MASTRANGELO, S. V. R., and ASTON, J. G., 1951, J. Chem.
Phys., 19, 1370
- MENDOZA, M. A., 1948, "Les Phénomènes Cryomagnétiques"
Hommage National À Paul Langevin et Jean Perrin,
Collège de France, Paris, p.53
- MEYER, H., 1959, J. Phys. Chem. Solids, 9, 296
- MEYER, L., 1956, Phys. Rev., 103, 1593
- NESTER, R. G., 1957, Rev. Sci. Instr., 28, 577
- NICOL, J., and SOLLER, T., 1957, Bull. Am. Phys. Soc.,
Ser II, 2, 63
- PEARCE, D. C., MARKHAM, A. H., and DILLINGER, J. R.,
1956, Rev. Sci. Instr., 27, 240
- PIPPARD, A. B., 1957, Elements of Classical Thermo-
dynamics, Ch. 9, Cambridge Univ. Press
- ROLLIN, B. V., 1936, Proc. VII Internat. Congress of
Refrig., 1, 187
- SCHAEFFER, W. D., SMITH, W. R., and WENDELL, C. B., 1949,
J. Chem. Soc., 71, 863
- SCHIFF, L., 1941, Phys. Rev., 59, 839
- SCOTT, R. B., 1959, Cryogenic Engineering, D. Van
Nostrand & Co., Ch. VI
- SHOENBERG, D., 1951, Superconductivity, Cambridge Univ. Press
- SINGH, R. P., and BAND, W., 1955, J. Phys. Chem.,
59, 663
- SOMMERS, H. S. 1952, Phys. Rev., 88, 113

- STEELE, W. A., 1956, J. Chem. Phys., 25, 819
- STRAUSS, A. J., 1952, Thesis, Chicago
- STRAUSS, A. J., 1953 (see Long and Meyer, 1953)
- TJERKSTRA, HOOFTMAN, and VON DER MEYDENBERG, 1953,
Physica, 19, 935
- WHITE, D., CHOU, C., and JOHNSTON, H. L., 1952, J. Phys.
Chem., 20, 1819
- WHITE, G. K., 1955, Can. J. Phys., 33, 119
- WHITE, G. K., 1959, Experimental Techniques in Low Temperature
Physics, Clarendon Press, Oxford
- UBBINK, J. B. and DE HAAS, W. J., 1943, Physica, 10, 465
- ZEMANSKY, M. W., 1957, Heat and Thermodynamics, 4th ed., Ch. 16
- BERMAN, R., FOSTER, E. L., ROSENBERG, H. M., 1955,
Brit. J. Appl. Phys., 6, 181

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